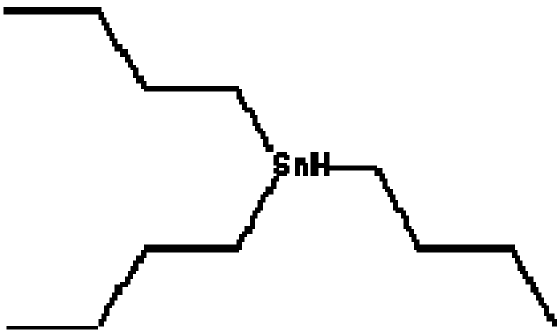
Tributyltin pollution on a global scale. An overview of relevant and recent research: impacts and issues.



Report to Dr. Simon Walmsley WWF UK. Godalming, Surrey.
Contract No: FND053998

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Executive Summary

The attachment of organisms to vessels or man-made structures in the aquatic environment is known as biofouling and can result in considerably increased maintenance and fuel bills. For this reason numerous attempts at control of biofouling were developed until the 1960s when organotin compounds such as tributyltin oxide (TBTO) were found to be highly effective. TBT was widely available in the United States and Canada by the late 1960s, but was not generally distributed in the UK and other countries until the early 1970s. TBT antifouling works by providing an unstable surface in which a toxic biocide is contained. Settling organisms are both unable to attach for prolonged periods and are poisoned by the organotin content.

Initially developed as free-association paint, TBT used contact leaching to release the biocide when exposed to water. However, the release rate was unpredictable and inconsistent in time and self-polishing co-polymer paints were subsequently developed. These use a polymer base through which the biocide discharge rate is regulated by reacting with water. The compound is slowly released as a result of wave action or forward motion. Once the surface covering is worn, biocide release begins again with the next layer and thus toxicity of the paint is consistent throughout. TBT paints extended the useful life of antifouling coatings, with ships able to continue commercial operations for up to 60 months, with consequent economic advantages, although after three years of operation TBT-based paints also need hull-cleaning. TBT, as most other antifouling biocides, needs several additional booster biocides in the formulation to work well.

Negative aspects of TBT were suspected in the late 1960s when it was realised that the release of readily bioavailable organotin into aquatic environments was impacting non-target organisms. Warnings of TBT use close to shellfish farms were made, but application of TBT paints grew, so that by the late 1970s they were commonly used on commercial and recreational craft from industrialised nations.

In the late 1970s and early 1980s, oyster (*Crassostrea gigas*) crops in Arcachon Bay, France, failed. Subsequent research identified that TBT had caused decreased spatfall and unnatural shell thickening; this was also observed in UK oysters stocks. The French Government imposed partial legislation against TBT, banning its use on recreational vessels less than 25 m long. Coinciding with this period, work in the UK showed that TBT was an endocrine disruptor in a marine stenoglossan gastropod species (*Nucella lapillus*) causing masculinisation (imposex) in females and widespread population decline. The sensitivity of this and other marine gastropods world-wide has led to their use as bioindicators of TBT pollution. The widespread recognition of effects promoted the spread of legislation and by the early 1990s, many nations had partial TBT bans in place. Research over the last twenty-five years has highlighted that numerous other non-target organisms species may be adversely affected by TBT, including many offshore, sublittoral species. These range from plankton to cetaceans and there are implications for bioaccumulation of TBT to the human food chain. The sentinel impact of TBT, imposex in female gastropods, is caused by an increase in the male sexual hormone, testosterone when TBT interferes with their endocrine systems. This effect may not be limited to molluscs alone and may underlie reproduction impacts by affecting mating triggers. Body burdens of TBT tend to be unevenly distributed throughout the year, with an elevated concentration of TBT in sex organs at the onset of the breeding season.

TBT can bond to suspended material and is deposited in benthic sediments, where it can last unaltered for decades, particularly in anoxic conditions. TBT can also volatilise into the air and is known to precipitate with rain. There are numerous estimates of how long

TBT will remain in sediments. These range from 7 to 30 years indicating that this 'reservoir' of the compound will require management for a considerable period; recent research in Arcachon Bay (one of the first TBT hotspots to be identified) has shown that sediment contamination was still affecting non-target species after 20 years. In 1991 offshore species found to be affected by TBT were close to merchant shipping routes and had not at all recovered around the turn of the century. Coastal TBT pollution has declined along many shores where legislation was put in place in the 1980s-1990s. For example, along the south coast of England and around oil terminals in the north of Scotland *Nucella lapillus* has recovered in many areas where, previously, pollution was severe. Nevertheless, apart from offshore waters, TBT pollution remains a concern in several coastal locations, in that impacted organisms have not recovered and levels in water still exceed the environmental quality target of 2 ngl^{-1}. Illegal use of TBT also remains a concern.

TBT 'hot-spots' are normally associated with commercial ports and dockyards (e.g. parts of Tokyo Bay, Japan; the Baltic; Southampton Water, UK; marinas on the coast of Israel) and the disposal of TBT contaminated sediments and dredged material from many of these areas is giving cause for concern. In addition, high levels can still be found around some marinas even though legislation is in place (e.g. Hamble River, UK; parts of the Algarve). Certain nations that have recently joined the EU may have hot-spots at both recreational craft and commercial ports (e.g. Polish Baltic ports) whilst there are other nations which have no TBT legislation in place: These include Israel, where, close to marina developments, sensitive female gastropods have developed imposex because of organotin-related endocrine disruption and far eastern countries where, for example, recreational boating around the island of Phuket is affecting gastropod populations. The findings of more than twenty affected non-target species in offshore South-East Asian seas in 1996, together with similar offshore impacts in the North Sea triggered the recognition by the International Maritime Organisation that TBT from merchant shipping posed a serious problem, and set the stage for the development of the antifouling convention (2001).

Contamination of sediments and the threat caused by continued use of TBT on commercial craft has long been recognised in many developed regions. Japan introduced stringent legislation against TBT in the early 1990s and was among the first nations to call for a world-wide ban. However, in much of Asia and particularly the rapidly developing economies of India and China, possible impacts on non-target species have increasingly become an area of concern. Contrary to global trends, in some Asian nations TBT levels and associated adverse effects are reported to be increasing. As there is a considerable dependence on mariculture in these regions, there are also concerns for TBT impacts on human health. Developing nations with quickly growing economies, but no TBT legislation may merit particular attention from future TBT research efforts.

The use of TBT as biocide for aquatic species was originally developed to combat the role of freshwater snails in transmitting schistosomiasis in Africa. Whilst freshwater ecosystems have not been the focus of as much research as marine systems, some landlocked nations have restricted the use of TBT to prevent leaching to wider aquatic habitats and impacts on non-target freshwater species.

After TBT was shown to cause imposex in non-coastal snails it became clear that the presence of TBT was ubiquitous in the world's oceans and present in a wide range of animals and plants. Among the most noteworthy findings of TBT in recent years is the presence of organotins in oceanic species, including cetaceans and pelagic fish. Global monitoring of TBT is now feasible using skipjack tuna (*Katsuwonus pelanus*) with results showing ubiquitous presence of TBT and highlighting areas that may have particularly high contamination (e.g. off the coast of Japan).

Marine pollution does not recognise national, regional or conservation boundaries; thus impacts from TBT can be seen or anticipated even in protected areas. Such concerns have been recorded in Europe (Italy) and further afield where for example port development has been proposed in a World Heritage Site in Vietnam. Closer to home there are indications that designated European Marine Sites are not immune from such pressures: for example whilst the intertidal gastropod *Nucella lapillus* has begun to recover on the north coast of the Isle of Wight (UK), on the south coast populations within the South Wight Marine Special Area of Conservation continue to be impacted.

Legislation to ban TBT globally was finally agreed at the 43rd meeting of the Marine Environmental Protection Committee (International Maritime Organisation, London) in 1999. Regulation of antifouling paints (with specific reference to TBT) was recommended with the last date for the application of organotin paints on vessels set as the 1st January 2003 and a total phase out of organotin antifouling coatings by 1st January 2008. Ratification of this legislation was slow and the January 2003 deadline was not met. Pressure on major shipping nations has continued, particularly from NGOs such as WWF, recently the percentage of shipping nations that have ratified the legislation has increased. However, there is still some way to go and whilst the legislation will require compliance from many developed nations, much of the developing world will not be signatories. Whilst nations have the right to seek economic growth they should at the same time heed the lessons of TBT impacts experienced by others and be mindful of requirements for legislation to protect the environment.

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TBT History, Legislation and Continuing Impacts World-wide

1. Introduction

Extensively used world-wide, organotin antifouling paints have proven very effective at inhibiting fouling organisms on ships and stationary structures. Unfortunately these tributyltin based paints have also achieved notoriety as highly toxic compounds adversely affecting non-target organisms throughout the world. These have ranged from plankton to larger cetaceans, although stenoglossan gastropods have been found to be the most sensitive family and globally have been used as biomonitors for TBT levels. Since the 1980s, biological and chemical testing has shown that in some areas TBT levels in water have declined - particularly where bans on its use on recreational vessels are in place.

However, where TBT can still be used on commercial craft there may be "hotspots" of contamination and levels in offshore seas remain high. High TBT levels can also be associated with marine sediments where it can remain for long periods, leaching out slowly unless disturbed by, for example, dredging. TBT pollution has been recognised as a global issue and there are plans for its total ban. Some argue that this is not necessary as (a) the problem may already be declining in many areas, and (b) the economic benefits of TBT antifouling (reduced fuelling costs and maintenance) outweigh the environmental disadvantages, although recent studies now show that other antifouling systems are at least as effective as TBT-based paints.

The International Maritime Organisation (IMO) proposed a total TBT ban from 2003 to be fully in place 2008. There has been reluctance to take this up, including by the UK, but ratification is increasing. However, even if IMO members take up the ban, there are areas in the world where nations may not become signatories and where restrictions on the use of TBT are non-existent or ineffective. This has continuing implications for local and global marine fauna including food resources, particularly in nations where economies are developing. Even in countries where TBT is banned, the legacy of the compound may persist for a considerable period. The current review summarises recent global research into TBT impact, discusses trends in relation to calls for a world-wide ban and considers the ongoing issues for marine conservation.

1.1 Biofouling and its prevention

Marine organisms have evolved to exist in the environmental extremes of intertidal, subtidal and pelagic habitats. For many, the ability to fix to substrata and remain attached in extreme weather has impinged on marine transport and other industrial activities; organisms affix to man-made structures such as vessels, oilrigs and pipelines and result in 'biofouling' (Clare, 1995; Callow and Callow, 2000; Yan and Yan, 2003; Konstantinou and Albanis, 2004). This often leads to a rougher surface, increased drag and higher fuel bills for shipping and heightened corrosion of stationary structures (Evans, 1981; Edyvean *et al*., 1985; Ten Hallers-Tjabbes, 1997); these effects are compounded by increased maintenance costs (Ludgate, 1987; Champ, 2001a, 2003). It has been shown that an estimated 50% of the operating costs of commercial vessels are from fuel consumption. A 10 μm increase in hull roughness is predicted to result in a 0.3-1% increase in fuel consumption and a hull with 5% fouling cover will result in a fuel consumption increase of 5-10% (Champ and Lowenstein, 1987); 33% fouling cover on a vessel will lead to 50% increased fuel costs (Ludgate, 1987). This coupled with regular dry-docking to remove biofouling organisms leads to increased shipping costs and decreased profit margins (Champ, 2001; Champ, 2003).

The International Maritime Organisation (IMO) promotes "*..efficiency of navigation and prevention and control of marine pollution from ships*" (IMO, 2000) and recognises that fouling on shipping is of major economic importance (Table 1.1). The impact of fouling organisms has, in recent decades stimulated the development of ever-more effective antifouling paints (e.g. Drouin, 2004; Xu *et al*., 2005) that minimise biofouling, thus reducing transport costs. In addition the role that biofouling can have as a vector in transport of alien invasive species may be prevented by alternative antifouling systems as successfully, or better than TBT. Despite this, fouling is still acknowledged to have a major impact on the profitability of commercial fleets and research into efficient antifouling coatings remains active (e.g. Konstantinou and Albanis, 2004; Yebra *et al.,* 2004). However, it is now acknowledged world-wide, that diffuse pollution released from antifouling compounds has had significant impacts upon non-target marine organisms (e.g. Pereira *et al.,* 1999; Konstantinou and Albanis, 2004; Roepke *et al*. 2005; Ruiz *et al*. 2005; Shim *et al*. 2005).

TABLE 1.1 BIOFOULING ISSUES: THE SHIPPING INDUSTRY PERSPECTIVE.
(Modified from IMO, 1999 based on MEPC, 1996).

Fouling explained	
What is fouling?	Fouling is an unwanted growth of biological material - such as barnacles and algae - on a surface immersed in water.
How much fouling does an unprotected structure get?	Vessel bottoms not protected by antifouling systems may gather 150 kg of fouling per square metre in less than six months of being at sea. On a very large crude oil carrier with 40,000 square metre underwater areas, this would add up to 6,000 tonnes of fouling.
Why do ships and static structures need antifouling systems?	Small amounts of fouling can lead to an increase of vessel fuel consumption, as the resistance to movement will be increased. A clean ship can sail faster and with less energy. Stationary structures suffer increased corrosion and therefore maintenance costs increase.
How do antifouling systems save ship owners money?	An effective antifouling system can save a ship owner money in a number of ways: • Direct fuel savings by keeping the hull free of fouling organisms; • Extended dry-docking interval, when the antifouling system remains viable; • Increased vessel availability - as it does not have to spend so much time in dry dock.

Over 2000 marine species cause biofouling (Evans, 1970), and of these, barnacles are the most significant (Christie and Dalley, 1987). Early records of fouling control showed that lead sheets (Stebbing, 1985), lime or oil laced with sulphur, arsenic and gunpowder (Clare, 1995; Ten Hallers-Tjabbes, 1997), and, latterly, copper sheeting (Stebbing, 1985), have been used. Success with copper sheeting led to the development of the first antifouling paints at the turn of the 19th century (Stebbing, 1985).

Many modern antifouling paints are based on bioavailable organometal compounds, where metal ions are bound with e.g. methyl, phenyl or butyl groups, producing toxic biocides which cause the death of fouling organisms at the settlement stage (Evans, 1970; Stebbing, 1985; Christie and Dalley, 1987). Original versions of organometal antifouling paints were based on copper (II) oxide and later arsenic and mercury (Stebbing, 1985; Christie and Dalley, 1987). In the 1960s, however, early versions of organotin paints were found to be more successful and cost-effective (Evans, 1970). As an antifoulant, the most effective organotin compounds proved to be based on tributyltin (TBT) and, to a lesser extent, triphenyltin (TPT). Although TBT was available in the United States by the late 1960s, it was not widely distributed in the UK and other countries until the early 1970s (Stebbing, 1985; Santillo *et al.,* 2002).

TBT was initially developed as a 'free-association' paint which used 'contact leaching' to release the biocide (Champ and Pugh, 1987); the TBT within the coating leaches out readily when it is exposed to water (Stebbing, 1985). However, the initial biocide release rate was subsequently found to be unpredictable so, in the 1970s, self-polishing co-polymer paints providing a constant and predictable release rate were developed. With co-polymer products the organotin compounds are chemically bonded to a polymer base (Evans, 1970; Champ and Pugh, 1987). The biocide discharge rate is regulated through the reaction of seawater with the surface of the compound; the biocide is slowly released by wave action or forward motion through water, which wears the co-polymer paint away. Known as self-polishing, once the surface covering is worn off, biocide release begins again with the next layer (Fig. 1.1). In this way the leaching rate and toxicity is consistent throughout the life of the paint. Such TBT paints extended the useful life of antifouling coatings, with ships able to continue commercial operations for up to 60 months without repainting (Callow, 1990).

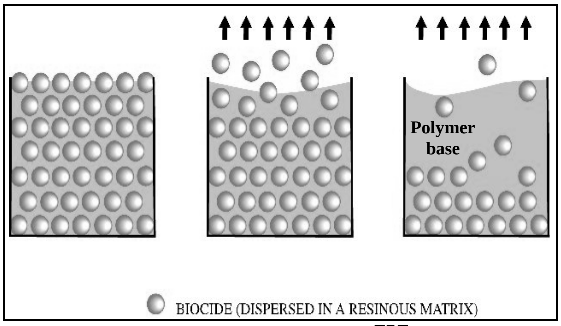

FIGURE 1.1 SCHEMATIC REPRESENTATION OF THE SELF-POLISHING ACTION OF TBT PAINTS.
(Modified from ORTEPA, 1999).

Because of the self-polishing nature of the TBT biocide system, not only were fouling organisms inhibited at an early stage, but also they were unable to attach for prolonged periods due to the unstable substrate. The result was the considerable reduction of drag-inflated fuel bills for shipping and a reduction in stationary structure corrosion (Ludgate, 1987; Clare, 1995). However, although TBT-based paints delivered an efficient solution to biofouling, concerns were raised over the indiscriminate release of the biocide into the marine environment; *i.e.* through self-polishing, the release of TBT from ships and other structures resulted in organotins becoming a diffuse pollutant.

1.2 Effects and sources of TBT

In Canada in the late 1960s, a recommendation was made that TBT use near shellfish farms should be avoided (Thomas, 1969; *op cit* Spence, 1989). In the 1970s and 1980s, an increasing number of potential links between the toxicity of TBT and adverse effects on non-target organisms were found (e.g. Good *et al*., 1980; Smith, 1981; Fridberg, 1985; Alzieu *et al*., 1986; Bryan *et al*., 1986; Wade *et al*., 1988). By the time that TBT antifouling paints were readily available in the UK it was apparent that there were associated problems. Despite this, their efficiency and wide availability in the 1970s resulted in their burgeoning use in the USA, Canada, and Europe (Evans, 1970; Champ and Pugh, 1987; Santillo *et al*., 2002).

From the early 1980s to the present day, studies have continued to show that at low levels organotin compounds impact on non-target species (e.g. Waldock and Thain, 1983; Alzieu *et al*., 1986; Bryan *et al*., 1986; Langston *et al*., 1990; Minchin and Minchin, 1997; Swennen *et al*., 1997; Stronkhorst *et al*., 1999;Yan and Yan, 2003; Terlizzi *et al*., 2004; Roepke *et al*., 2005). This includes marine microorganisms such as phyto- and zooplankton. For example, Beaumont and Newman (1986) found that at levels in the range 0.001-1.092 μgl^{-1} TBT restricted the growth of micro algae and U'ren (1983) found that the substance was highly toxic to the copepod *Acartia tonsa*. Dahl and Blanck (1996) also reported that artificial periphyton communities suffered reduced biomass and photosynthesis and showed a shift towards tolerant species at TBT levels of 1 ngl^{-1}.

In the UK and elsewhere, following the introduction of legislation partially banning the use of TBT, restrictions on the use of organotin antifouling paints have stemmed inputs from the leisure fleet. The highest levels of TBT are now generally associated with commercial shipping movements and shipping maintenance sites (e.g. Minchin *et al*., 1997; Morgan *et al*., 1998). Langston *et al*., (1994) reported that waterborne TBT at Lymington near Southampton (UK), used largely by pleasure craft, had declined to ~ 7 ngl^{-1} in the early 1990s - a marked decrease from a high 800 ngl^{-1} in 1988. In comparison, at the UK commercial port of Southampton, concentrations in June 1994 were still recorded at 43 ngl^{-1} and work by Thomas *et al*., (2001) found TBT levels near the commercial docks at 33 ngl^{-1} and even in the nearby recreational harbour of Hythe marina levels were still at 14 ngl^{-1}. TBT at these levels affects the physiology, growth and behaviour of many organisms. World-wide organotin pollution has caused widespread death in several commercial and non-commercial species (e.g. U'ren, 1983, Alzieu *et al*., 1986; Bryan *et al*., 1986; Gooding *et al*., 1999; Jha *et al*., 2000; Zhou *et al*., 2001; Birchenough *et al*., 2002; Kobayashi and Okamura, 2002) and where commercial shipping is prevalent this situation continues (e.g. ten Hallers-Tjabbes *et al*., 1994, 1996, 2003[a]) including offshore species continuing to be affected at the turn of the millennium (Ten Hallers *et al*., 2003[a,b]; Santos et al., 2002).

Antifouling paints were developed to inhibit barnacles in particular, but paradoxically they are among the least affected by TBT (Goldberg, 1986). TBT is poisonous to a range of organisms from plankton (U'ren, 1983) to higher-level organisms (e.g. Tanabe *et al*., 1998) and potentially humans (Heidrich *et al*., 2001; Nielsen and Rasmussen, 2004). However, TBT is most harmful to stenoglossan gastropods, by virtue of its effects on reproduction, with at least 72 species known to be susceptible, world-wide (Morgan *et al*., 1998; see also Ellis and Pattisina, 1990). TBT interrupts stenoglossan gastropod endocrine systems by inhibiting the P450 cytochrome (aromatase) molecule (Spooner *et al*., 1991). The molecule is responsible for converting androgens (which have male hormone properties) to oestrogens. Molluscs are dependent on P450 and due to slow reaction rates cannot metabolise TBT sufficiently rapidly to detoxify its effect on their endocrine systems; thus masculinisation occurs in females. This effect may not be limited to molluscs alone and may underlie the impact on reproduction by affecting the trigger to mate (Straw and Rittschoff, 2004). The body burden of TBT tends to be unevenly distributed throughout the year, with an elevated concentration of TBT in the sexual organs at the onset of the breeding season (Mensink *et al*., 2001).

Although TBT impacts became apparent in the late 1970s and 1980s, restrictions were not put in place until it was confirmed that commercial shellfish stocks were being affected (Alzieu *et al*., 1986; Waldock, 1986). The commercial oyster (*Crassostrea gigas*) industry in Arcachon Bay, France, declined at the same time as growers of *C. gigas* along the east coast of England reported abnormal shell forms (Alzieu *et al*., 1986; Alzieu, 1991). A link to TBT was made by researchers in the UK (Waldock and Thain, 1983; Thain and Waldock, 1986) and France (Alzieu *et al*., 1986). In adult *C. gigas,* exposure to levels of 20 ng l^{-1} TBT, and perhaps as low as 2 ng l^{-1}, causes shell deformities (excessive shell calcification resulting in reduced inter-valve volume). From 1975, this deformation, coupled with dramatic reduction in spatfall, was recorded in French oyster culturing areas (Alzieu *et al*., 1986). *In situ* experimentation showed that embryogenesis and larval development of oysters had been inhibited by TBT (Alzieu, 1995; Alzieu, 2000). Subsequent work on *C. gigas* in the River Crouch, Essex, revealed that shell deformities were also occurring within this population (Waldock and Thain, 1983; Thain and Waldock, 1986; Waldock, 1986) and TBT levels of up to 400 ngl^{-1} were found (Waldock, 1996). The financial loss to the oyster industry from 1975-1982 totalled some $150 million

(Langston, 1995). In January 1982, the French Government banned the use of TBT-based paints on all vessels under 25m length overall (LOA). This was followed by similar legislation in the UK and USA (Champ, 2000). The effect of the ban soon became apparent, with tin levels in oysters from Arcachon Bay falling from 3.40 µg g^{-1} dry weight in August 1982, to 0.50 µg g^{-1} by August 1984 (Alzieu *et al.*, 1986). However, TBT remained in use on larger vessels on the assumption that levels in offshore seas would be too low to cause effects. This claim has been challenged by findings of imposex in offshore snails in the early 1990s where none had been found in the early 1970s (Ten Hallers-Tjabbes *et al.*, 1996).

Although legislation was put in place, waterborne TBT levels were slow to improve in some areas, perhaps because of slow release of organotin bound up in sediments (Langston *et al.*, 1990; Langston and Burt, 1991; St. Jean *et al.*, 1999). TBT partitions between sediment and water at ratios of several thousands to one (Langston and Burt, 1991; Ruiz *et al.*, 1996) Whilst TBT in water was disappearing with a half-life of some three years after legislation, in contrast, in sediments TBT was far more persistent. These estimates of persistence vary from upwards of 7 years (Langston *et al.*, 1994), to 10 years (Dowson *et al.*, 1996) or as suggested by Macguire (2000) "*there may be a legacy problem* [of TBT] *in sediments in some locations in Canada for perhaps 20 to 30 years after a total ban*" (see also Bryan and Langston, 1992).

The persistence TBT in the water column and sediments is dependent on a combination of hydrodynamic and biogeochemical factors (Ruiz *et al.*, 1996). Hydrodynamics such as localised circulation patterns and gyres can determine the time that pollutants remain in the water column. TBT partitioning is affected by salinity and the compound can accumulate at the air-water interface. Dissolved TBT can also volatilise into the air (Amoureux *et al.*, 2000) (and be deposited through rain (Ariese *et al.*, 1997)) while stratification of the water column can affect the vertical distribution of TBT (Ten Hallers *et al.*, 2003). TBT breakdown and loss to the atmosphere may be stimulated through photolysis (Ruiz *et al.*, 1996). However, TBT also bonds to suspended particulate matter (SPM) and is deposited on the benthic (bottom) sediment layer (see Fig. 1.2 for a summary of the process). The net result of long-term residence of TBT in sediment reservoirs and subsequent leaching into the water column may result in sensitive non-target organisms being affected for many years. The degradation of TBT is temperature dependent and becomes very slow in cold climates and vulnerable polar areas.

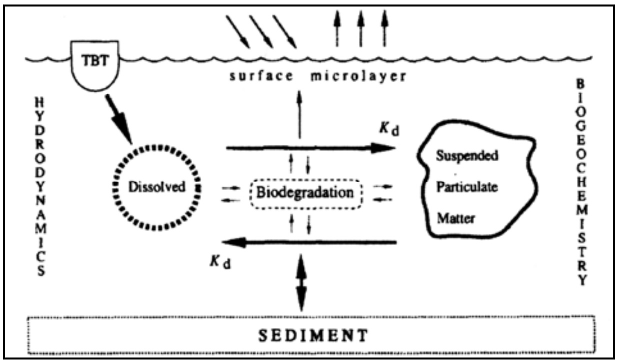

FIGURE 1.2 THE FATE OF TRIBUTYLTIN DUE TO DEPOSITION, HYDRODYNAMICS AND PHOTOLYSIS IN THE COASTAL MARINE ENVIRONMENT.
(Where kd is partition coefficient - influenced by pH, dissolved organic matter and SPM (load, size and organic content)) (Ruiz *et al.*, 1996).

There is also increasing concern over the release of TBT from sediments through dredging or similar disturbance (Brack, 2002) and difficulties in managing this problem have frequently been highlighted (e.g. Svavarrson, 2001; Santos *et al.,* 2004). Maritime ports increasingly have to expand the throughput of containerised transport vessels to meet competition. Throughout Europe there are plans to increase shipping by turning ports into regional hubs (Marcadon, 1999) which can include proposals for developments near to sensitive marine habitats, (e.g. the recent (unsuccessful) proposal for Southampton Docks, UK, Adams Hendry, 2000). Such activities could lead to further recharging of sediments with TBT. In addition, as deeper channels may have to be maintained to greater depths to potentially allow access to larger vessels (Side and Jowitt, 2002), the impact from dumped spoil and resuspension events may increase at sites not yet widely affected by TBT (Svavarrson, 2001). The management of dumped dredge spoil is a widespread concern, as resuspended material containing TBT can potentially be transported extensively including into statutory conservation areas. Such designations themselves do little to defend protected areas from transported pollutants (Boersma and Parrish, 1999; Terlizzi *et al.,* 2004). There is currently a need to establish cause-effect relationships between dredged spoil disposal activities and the impacts of TBT on protected organisms in selected ecosystems, together with the potential influence of illegal use (Kettle, 2000). Without such trials, 'knee-jerk' reactions and lack of real evidence may lead to poor decisions (Champ, 2003), either failing to protect vulnerable species, or, if legislation controlling the disposal of dredge spoil were tightened, unnecessary increases in disposal costs.

2. Legislation

The collapse of the oyster industry in Arcachon Bay, France, resulted in restrictions upon the use of TBT (Alzieu *et al*., 1986, 1991). In January 1982, the Ministry of the Environment for France enforced a 2-year ban on paints containing more than 3% by weight of TBT (Champ, 2000). Initially the ban was for vessels less than 25 tonnes in weight and only applied to Atlantic coasts and the English Channel. In September of the same year the ban was expanded to cover all the coastal areas of France and all TBT paints. The ban was also extended so that TBT paints could only be applied to vessels over 25 m length over all (LOA). An exception was made for aluminium hulls, as copper based paints, the main alternatives to TBT, caused their corrosion (Lewis, 1999). TBT-based paints also use copper as a hydrolysing agent in the formulation, with comparable levels to copper-based antifouling compounds.

UK legislation was introduced in July 1985. The Environment Minister set up actions for the control of TBT, comprising methods to control the sale of the most damaging TBT paints (i.e. free-association) and limiting the tin content in co-polymer paints; a notification scheme for all new antifouling sales agents; guidelines for removal of old and painting on new antifouling coatings; development of a provisional Environmental Quality Target (EQT) for TBT in water at 20 ngl^{-1} and the co-ordination of research and monitoring schemes to enable the UK Government to assess the efficacy of legislation put in place (Morgan, 1998; Champ, 2000; Santillo *et al*., 2002). Following these proposals, on the 13th January 1986, the UK regulations were enforced under the Food and Environment Protection Act 1985, Part III. Consequently, TBT content was restricted to 7.5% in copolymer paints and 2.5% in free association paints. Further to the above measures, on the 1st July 1987, UK legislation banned the use of TBT on vessels less than 25 m LOA (Matthiessen *et al*., 1995; Champ, 2000). In addition, the use of TBT was banned on aquaculture cages (Balls, 1987; Champ, 2000). Finally, in 1987, the EQT for water-borne TBT was lowered to 2 ngl^{-1} and replaced by an Environmental Quality Standard (EQS). The last was act was to ban the retail sale of TBT in May 1987 (Champ, 2000): currently

(2006) UK TBT legislation remains unchanged from that put in place in 1987 though the UK will be bound to comply with EU agreement to adopt the directive banning TBT in 2008.

In the United States, there was initial reluctance to legislate against TBT and the US Navy considered that the problems occurring in Europe were because of the excessive use of free association paints on small vessels. They were hesitant to enforce legislation against TBT as they estimated that the benefit of its use was around \$100-130 million per annum (Champ, 2000). By mid 1987, however, coastal USA states were considering TBT restrictions. These were initially at State rather than Federal level with allowable waterborne limits ranging from 10 ngl^{-1} for Virginia (which shortly after changed to 1 ngl^{-1}), to 6 ngl^{-1} in California (Champ, 2000). Following differing state versions of TBT legislation, the Federal Government enacted the Organotin Antifouling Paint Control Act of 1988. Similarly to the UK, TBT was banned on vessels less than 25 m LOA and release rates were set (Ten Hallers-Tjabbes, 1997; Champ, 2000).

Controls introduced in Canada, Australia and New Zealand during 1989 were similar to those in the USA (Ten Hallers-Tjabbes, 1997). Monitoring in Japan found 'biologically significant' levels of organotin in the local marine environment and strict controls were put in place for the use of TBT paints on non-aluminium vessels (Champ, 2000). The legislation amounted to a ban of TBT on new hulls, and on its manufacture, use and import (Champ, 2000). Legislation against TBT has subsequently been enacted in many countries around the world, including some with no direct link to the sea. Austria and Switzerland banned TBT in their lakes and rivers, thus minimising direct effects on freshwater communities and indirect impacts on marine habitats downstream (e.g. Becker Van-Slooten. and Tarradellas, 1994; Stab *et al*., 1996; Day *et al*., 1998; see Fent, 1996 for review). By 1992 numerous nations had placed restrictions on the use of TBT (Table 1.2).

Amongst its many remits the International Maritime Organisation (IMO) provides an international focal point for the monitoring and introduction of policy aims regarding TBT use on commercial vessels. Within the IMO the formation of policies to safeguard marine habitats is considered within the Marine Environmental Protection Committee (MEPC). The MEPC consists of signatory states to the International Convention for the Prevention of Pollution from Ships (MARPOL 73/78, 1973).

In 1990 the MEPC agreed resolution MEPC 29 that recommended the promotion of methods controlling and minimising the adverse affects of TBT, which ensured that users and producers of TBT were registered and regulated. The resolution included the stipulation that part of the profits from TBT sales be put towards research into alternatives. In addition an 'environmental degradation fund' was created for research into TBT in the marine environment. The resolution agreed the international banning of TBT on vessels less than 25 m LOA and a vessel release rate for TBT/water of 4 $\mu g\ cm^{-2}$ per day (Ten Hallers-Tjabes, 1997; Champ, 1999, 2000). The legislation was taken up internationally with governments imposing national and local regulations. Following the recommendations of MEPC29, Japan called for the world-wide ban on TBT at the 30th MEPC meeting in 1990. Research had found biologically significant levels of TBT in the marine environment and it was suggested that the high incidence of ocean going vessels passing through Japanese waters was to blame (Champ, 1999).

In November 1998 at MEPC 42, several nations (Belgium, Denmark, France, Norway, the Netherlands, Sweden and the UK) called for a world-wide ban on TBT paints (MEPC, 1998; Champ, 2000). At the MEPC 43rd session (June to July 1999) it was recommended that legislation be passed to regulate antifouling paints with specific reference to phasing out organotin compounds (Champ, 2000). The MEPC subsequently suggested a 10-year period to implement the planned ban. The last allowable date for the application of organotin paints on vessels being set as the 1st January 2003 and a total phase out of organotin antifouling coatings by 1st January 2008 (Champ, 2000).

TABLE 1.2 REGULATIONS CONTROLLING THE USE OF TBT PAINTS WORLD-WIDE BY COUNTRY UP TO 1992 AND IMO PROPOSALS FOR 2003 AND 2008.
(Modified from Ortepa, 1999).

Europe	Year	Regulations
Austria	NA	Banned the use of TBT antifouling paint in fresh water lakes.
Europe (EC states)	1991	Prohibited the use of TBT-based paints on vessels less than 25 m LOA. TBT antifoulants available only in 20 L containers.
Europe (non-EC)	Vary	Prohibited use of TBT-based paints on vessels less than 25 m (most states).
Finland	1991	Prohibited the use of TBT-based paints on boats less than 25 m LOA.
France	1982	Prohibited the use of TBT-based paints on vessels less than 25 m LOA, except for aluminium-hulled vessels.
Germany	1990	Prohibited the use of TBT-based paints on vessels less than 25 m LOA. Ban on retail sale. Ban on its use on structures for mariculture. Regulation for the safe disposal of antifouling paints after removal.
The Netherlands	1990	Prohibited the use of TBT-based paints on vessels less than 25 m LOA Washing/blasting slurry used to prepare TBT antifoulants may be treated as hazardous waste TBT antifoulants available only in 20 L containers All antifoulants must be registered
Norway	1989	Prohibited the use of TBT-based paints on vessels less than 25 m LOA.
Sweden	1989 1992	Prohibited the use of TBT-based paints on vessels less than 25 m LOA. Maximum leaching rate of 4µg/cm2/day for vessels greater than 25 m LOA. All antifoulants must be registered.
Switzerland	1987	The use of TBT-based antifouling paints is banned in fresh water lakes. All antifoulants must be registered
United Kingdom	1985 1987	Sale of TBT-based paints restricted, effective bar on TBTO free-association paints. Prohibited the use of TBT-based paints on vessels less than 25 m LOA and on fish-farming equipment. TBT antifoulants available only in 20 L containers. All antifoulants to be registered as pesticides; Advisory Pesticides Committee must approve sale and use. Washing/blasting slurry treated as hazardous.
N. America	**Year**	**Regulations**
Canada	1989	Prohibited the use of TBT-based paints on vessels less than 25 m LOA, except for aluminium-hulled vessels. Maximum leaching rate set for vessels greater than 25 m in length All antifoulants must be registered
United States	1988 1990	Prohibited the use of TBT-based paints on vessels less than 25 m LOA, except for aluminium-hulled vessels. Maximum leaching rate of 4 µg/cm^2/day for vessels greater than 25 m in length. All antifoulants must be registered. TBT-based antifouling paints can only be applied by certified applicators.
S. h/sphere	**Year**	**Regulations**
Australia	1989	Prohibited the use of TBT-based paints on vessels less than 25 m) LOA. Maximum leaching rate of 5 micrograms per square centimetre per day (µg/cm2/day) for vessels greater than 25m LOA. All dry-docks must be registered with the Environmental Protection Agency because of discharges. All antifoulants must be registered.
New Zealand	1989 1993	The application of TBT copolymer antifouling paint is banned with three exceptions: hulls of aluminium vessels, the aluminium out-drive or any vessel greater than 25 m LOA. The application of TBTO free-association paints is banned. Maximum leaching rate of 5µg/cm^2/day for vessels greater than 25 m LOA. All antifoulants must be registered. Use of any organotin containing antifouling paint prohibited.
South Africa	1991	Prohibited the use of TBT-based paints on vessels less than 25 m LOA. TBT antifoulants available only in 20 L containers All antifoulants to be registered.
Hong Kong	NA	All TBT antifoulants must have a valid permit for import/supply. All antifoulants must be registered.
Japan	**Year**	**Regulations**
Japan	1990 1992	TBT banned for all new vessels. TBT banned for all vessels.
IMO measures	**Year**	**Regulations**
World-wide ban proposal	2003-2008	Proposed ban for 1st Jan 2003 – no reapplication of TBT. 1st Jan 2008 No ships or structures shall bear TBT. To be ratified if 25% of shipping tonnage or 25 of the worlds shipping nations sign.

The decision to ban TBT applications on the 1st January 2003 was contingent on the support of 25 states representing 25% of the world's merchant shipping tonnage (IMO, 1999). The 159 member states of the IMO accept the proposed ban in October 2001 (IMO, 2001). Unfortunately by July 2002 the treaty had not been validated by any of the initial signatory nations. Thus it was identified that the regulations could not be enforced until twelve months after the 25 state signatory figure had been reached (ENDS, 2002a).

Whilst waiting for the ban, two nations attempted to enforce their own legislation. Belgium in 2000 and Germany in 2001, with the backing of the Danish Ecological Council (Ege and Boye, 2001), attempted to adopt the proposed ban conditions; both were rejected. Belgium was told that it could not introduce the planned law because it had agreed to allow restricted use of TBT under a European Union directive in 1999. The European Commission ruled out new evidence from the Belgian authorities, that high TBT levels in flatfish increased the risk to human consumers, warranting tougher measures (ENDS, 2002a). The German application was rejected on similar grounds. Both nations were advised to wait for world-wide legislation being developed by the IMO.

In November 2002 the European Union's transport committee agreed that the IMO proposal to ban organotins from 2003 should be supported. The Commission noted that the draft regulation, agreed in 2001, was unlikely to take effect from the 1st January 2003 due to procedural delays, but that it would be implemented by the summer (ENDS, 2002b). IMO members, however, were reluctant to take up the ban and the deadline of 1st January 2003 passed. The second half of the IMO ban scheduled for 1st January 2008 is still in place. This states, "ships shall not bear such [TBT] compounds on their hulls or external parts or surfaces" (IMO, 2001). By April 2004 the uptake of the proposed ban amounted to 9.65% of the worlds shipping tonnage. By November 2004, however, this figure had reduced to 9.04% due to non-signatory nations registering greater shipping tonnage (IMO pers. com. 2004). Encouragingly the most recent communication with the IMO revealed that 17.27% of the worlds shipping tonnage was now covered by the agreement; largely due to the joining of Greece and Cyprus (IMO pers. com 2006). If the UK (worlds fifth largest shipping nation by tonnage) or Panama, for example, were to join, the ban may be fully ratified.

Some researchers have suggested that the TBT ban is unnecessary (e.g. Evans, 1999; Lewis, 1999) arguing that legislation in place is sufficient and that a ban is premature and unrealistic until viable TBT alternatives are found. In addition, several authors (Champ, 1999, 2000, Abbott *et al*., 2000; Abel 2000) have stated that TBT concentrations in water and sediment had declined to acceptable levels. The validity of research into the impacts of TBT has even been questioned by some workers who suggest that many conclusions are based on generalities (Champ, 2000). In the UK Evans *et al*., (2000[a]) reported that even though TBT impacts on the bioindicator species *Nucella lapillus* (the common dogwhelk) were marked, predicted population extinctions had not occurred and that dogwhelks were found on all of the sites surveyed. Champ (2000) has expressed doubts as to whether TBT is bioaccumulated through the food chain. In addition, Evans *et al*., (2000[b]) identified other causes of imposex in *N. lapillus* including the endocrine disruptor nonylphenol and parasitism by worms, thus questioning its status as a bioindicator for TBT.

At the time of the controversy over TBT, the MEPC were promoting research into alternatives (Champ, 2001b; Santos *et al*., 2002) and some were reported as adequate (Ten Hallers-Tjabbes, 1997). Abel (2000) highlighted, however that there were disincentives towards developing TBT alternatives since shipping operators funded much of the research. It was felt that if TBT were successfully banned without adequate alternatives the costs would be to shipping operators with the benefits going to paint manufacturers. Therefore shipping companies may have been reluctant to promote less effective and more costly alternatives (Abel, 2000). It was argued that the proposed TBT ban had been ill considered and that the effects of a total ban were potentially worse. As a result ships would travel more slowly, use more fossil fuels and a switch to less environmentally friendly modes of transport such as air and land may occur (Abel, 2000; Strandenes, 2000).

Batt (1999) summarised the issues that had led to what was considered by some to be a premature ban on TBT. These issues included the absence of proven alternatives to TBT; the fact that no specific criteria had been set to evaluate alternatives; and the fact that those preparations already available did not meet US VOC (volatile organic compounds) emissions restrictions. In addition, the concerns regarding economic consequences to the shipping industry resulting from the enforced use of TBT alternatives were seen as important (Ludgate, 1987; Champ, 2001a). Supporting some of the views expressed by Champ (2000), it was also felt that the decisions to ban TBT were based, partly, on flawed research (Batt, 1999).

Apart from concerns about increased fossil fuel use and a growth in the use of even less environmentally friendly modes of transport, there are also potential problems with the disposal of TBT residue after it has been removed from hulls. Abbott *et al*., (2000) commented that there is no provision in place for the safe disposal of TBT polluted residue after 2008, hence the excess may be dumped into sensitive areas in unregulated countries in the developing world (e.g. Greenpeace, 2000). Furthermore some advocates of TBT stated that alternatives were ill developed, less effective and had as much potential for insidious pollution (Batt, 1999; Abbot *et al*., 2000; Champ, 2001).

It would seem there are still strong lobbies both for and against the continuing use of TBT-based antifouling paints which only more appropriate, targeted research is likely to resolve.

3. Continued global effects of TBT on non-target species

3.1 Background

Following the introduction of TBT legislation over the last two decades, there have been some improvements in levels of TBT pollution, particularly at sites where recreational craft are dominant e.g. Iceland (Svavarsson, 2000; Jorundsdottir *et al*., 2005), Australia (Batley *et al*., 1992; Rees *et al*; 2001), United Kingdom (Waite *et al*., 1991; Evans *et al*., 1996; 1998; Hawkins *et al*., 2002). In some areas the proximity of gastropod bioindicator populations to TBT sources can be critical in determining recovery from organotin pollution. Harding *et al*., (1997) found that *N. lapillus* close to the Sullom Voe oil terminal in the Shetlands (UK) were still impacted by TBT, whilst populations a short distance away on more exposed shores showed imposex reductions. However, whilst improvements through recreational vessel restrictions are widely reported (except in some countries with no legislation), this should not distract attention from findings that show TBT and associated impacts levels are still high close to industrial shipping routes (see Ten Hallers-Tjabbes, and Boon, 1995; Ruiz, 1998; Fent, 2004; Santos *et al.,* 2004) and in locations where dredge spoil disposal has been shown to have an impact (Weichsel *et al*., 1998; Svavarsson *et al*., 2001; Santos *et al*., 2004). Where the ban on recreational craft has been implemented, it has been effective, however with regard to commercial ships, impacts continue to be reported in both regulated and unregulated nations (Ten Hallers-Tjabbes *et al*., 1994; Ide *et al*., 1997). Nevertheless, such impacts should be considered in conjunction with other potential anthropogenically derived effects (e.g. see Ten Hallers-Tjabbes *et al*., 1994; Strand and Jacobsen, 2002).

Some sites with continuing high organotin levels are considered to be hot-spots (e.g. Evans *et al*., 1995, 1998; Rilov *et al*., 2000; Birchenough *et al*., 2002; Gibson and Wilson, 2003).

In the UK and elsewhere these are normally close to industrial ports (e.g. Harding *et al*., 1997; Morgan *et al*., 1998; Miller *et al*., 1999; Horiguchi *et al*., 2004), but may also be related to dumped dredge spoils (e.g. Portugal Santos *et al*., 2004). In the absence of any effective legislation TBT may still be an issue even in small-boat dominated areas. Rilov *et al*., (2000) investigated imposex near two marinas in Israel, which has no current TBT legislation. They found that high organotin levels from the marinas caused imposex in two species of gastropod (*Stramonita haemastoma* and *Hexaplex trunculus*); imposex significantly decreased away from the sites. A monitoring programme, officially started in 2001 investigating TBT and other pollutants in port sediments, showed that concentrations were in the region of 100 μg/kg (Barak Herut *et al*., 2003). Thus even where legislation is in place, there are some areas where the legacy of TBT use produces great uncertainty over the prognosis and timescales for recovery. For example in the UK *N. lapillus* populations on the Isle of Wight continued to be affected after the 1987 legislation (Langston *et al*., 1997; Bray and Herbert, 1998; Herbert *et al*., 2000). There are impacted populations at sites on the south coast, which are unlikely to have direct shipping influence; the cause of this needs investigation (Bray and Herbert, 1998; Herbert *et al*., 2000).

Thus, whilst TBT pollution is reducing at many locations previously impacted by recreational craft (e.g. north coast of the Isle of Wight, UK), and at many sites close to commercial ports (e.g. Sullom Voe, Shetlands), and shipping routes world-wide there are still hot-spots, despite current legislation. In some cases, the TBT pollution, albeit at low levels, is unexpected due to remoteness from commercial vessels (e.g. Italian Marine Protected Areas - MPAS). The relevance of TBT occurrence at such sites, particularly those of high conservation value, has still to be addressed adequately.

3.2 Africa

In the early stages of the development of TBT as a biocide, TBT compounds were tested in Africa as molluscides against freshwater snails, which transported the disease schistosomiasis (WHO, 1990). This led to the development of TBTO as an antifouling compound and its subsequent commercial use in the 1960s. Ironically, research into TBT impacts on African coastal ecosystems is relatively limited. This perhaps reflects greater industrialisation and development on the northern side of the Mediterranean sea, where TBT and other pollutant (e.g. mercury) levels are perceived to be higher (Gabrielides, 2000). This does not rule out the occurrence of TBT pollution in the North African region; in the mid to late 1980s, research in Algeria showed that TBT was a factor in the decline of the mussel species *Perna perna* (Abada-Boudjema and Dauvin, 1995). There has been limited work carried out on TBT pollution in neighbouring Tunisia. Recent work, however, has shown significant TBT contamination of sediments and mussels in the Bizerte Lagoon (from industry and antifouling paint) and indicates the need for more research in the region (Mzoughi *et al*., 2005).

In contrast to much of the North African coastline, the major industrial port and marina location of Alexandria has been the focus of several TBT studies. Alexandria (and eastwards to Port Said), is a major staging post for commercial vessels plying the Suez Canal from and to the Arabian Gulf. Barakat *et al*., (2001) collected sediment samples from 23 locations in Alexandria harbour and found TBT concentrations ranging from 727-2,067 ng/g^{-1}. Biselli *et al*., (2000) found concentrations similar to this at marina sites in the Baltic and concluded that they represented an ecotoxicological risk (even eight years after a ban in the region). The TBT levels found in sediments and biota of Alexandria harbour (e.g. Abdallah, 1995; Mohamed, 2005) may take many years to decline and Barakat *et al*., (2001) concluded that the high TBT-sediment concentrations indicated that degradation processes to mono or dibutyltin were minor, probably due to the anoxic conditions at the sampling sites and/or relatively fresh input of TBT. Thus the potential for impact on local marine biota may persist for some years. There are records of gastropods *Thais carnifera* being severely impacted in the region of the Suez Canal (Hanafy, 1996) and further biomonitoring in the area of Egyptian waterways and ports would help determine the extent of the problem in this important bottleneck for commercial shipping.

The work on contaminated sediments around North African coasts indicates the potential for TBT impacts, or that such effects have previously occurred (e.g. Abada-Boudjema and Dauvin, 1995). South of the Sahara, Marshall and Rajkumar 2003 stated that "*no published reports of TBT contamination are known for any sub-Saharan African harbours*": however one report (Nyarko and Evans, 1997) investigated TBT levels in gastropods in Ghanaian ports (Tema and Takoradi) and found little evidence of organotin pollution. Marshall and Rajkumar (2003) investigated TBT levels in the South African harbours of Durban and Richards Bay using a gastropod bioindicator species and found variable levels of TBT impacts from severe to minimal, depending on localised vessel activity. There was also some concern that TBT pollution in the area may affect nursery fish grounds, which are regionally important. In addition the disposal of dredge spoil may be an issue for South Africa's numerous Marine Protected Areas (MPAs) such as the Aliwal Shoal, south of Durban, and Richards Bay. Dredge spoil disposal may require further investigation in these areas as it is dumped offshore (Jordaan, 1970). More recently, in relation to Durban and Port Richard harbour development, Forbes *et al*., (1996) stated that "*environmental impacts associated with engineering developments are becoming a significant factor in the decision making process*". Such impacts should include consideration of sediment disposal on MPAs.

3.3 Americas

North America saw some of the earliest research into imposex-affected gastropod species (e.g. Smith, 1971), and early work in Canada advised against the use of TBT close to shellfish areas (Thomas, 1969, *op cit* Spence, 1989). Recent research in North America has shown that despite partial federal legislation in 1988 and 1990 (Champ, 2000), TBT impacts continue to be recorded. In Canada, although recovery has been seen in some imposex affected gastropod populations (e.g. Victoria and Mission Point), *Nucella lima* populations were still absent at Vancouver and high TBT levels were found in the tissues of the mussel *Mytilus trossulus*. This was believed to be due to the continued use of TBT on vessels greater than 25 m long (Horiguchi *et al*., 2004). Watson (2003) has suggested that improvements in gastropod populations in the cities' harbour would be expected to match those outside if TBT use were phased out.

In the USA TBT pollution continues to affect marine communities around large ports where inputs may be due to industrial shipping or dry-docking activities (e.g. Peachey, 2003). In Mobile Bay, Alabama it was found that TBT levels in oyster tissues did not show a significant temporal decrease (Peachey, 2003). Elgethun *et al*., (2000) reported high, but decreasing, TBT-sediment concentrations (24-1196 $\mu g\ kg^{-1}$) for Oregon shipyards in 1997- similar to general trends of TBT decline in estuaries in the USA (Peachey, 2003). General improvements have been particularly evident in locations dominated by recreational craft (e.g. Lenwood *et al*., 2000), although there are some areas where recent TBT research is surprisingly limited or does not exist e.g. San Francisco Bay (Brown *et al*., 2003).

Panama may have the largest vessel throughput of any South or Central American country, however, recent research efforts on the impacts of TBT were again not apparent. As Godoi *et al*., (2003) point out, TBT impacts have mainly been studied in the Northern

Hemisphere in the past; although, with the growth of trade in Asia and India, TBT research may be beginning to change focus. There are some data along the Chilean coast (e.g. Gooding *et al*., 1999) and Argentina (Penchaszadeh *et al*., 2001) where high levels of TBT pollution were indicated. For Brazil, In São Paulo, Godoi *et al*., (2003) found TBT contaminated sediments at levels of 360 and 670 ng/g^{-1} at the two most contaminated sites. They concluded that TBT use on leisure boats and dry-dock activities needed further control. Impacts upon the local marine biota were not investigated, but at these levels effects of TBT pollution on sensitive species might be expected. In addition to the major city of São Paulo, Stringer *et al*., (2000) also reported TBT contaminated sediments from the Baia de Guanabara in Brazil including a reportedly "pristine" location. It was concluded that Brazilian Naval activity and docking were likely sources and Fernandz *et al*., (2002) found continued evidence of TBT impacts in the bay with some high imposex impacts in *Thais haemastoma* populations.

Legislation in Canada has been the subject of Special Review and the decision detailed that all TBT compounds would be phased out in 2002 (Special Review Decision SRD2002-1; Pest Management Regulation Agency, 2002). This was taken due to the "*unacceptable risk to the marine environment*" from TBT compounds. Thus it may be expected that TBT levels in affected areas will decline, although very high levels in sediment have been recorded (e.g. Maguire, 2000) and recovery may be a prolonged process. In the USA partial legislation against TBT was enforced in 1988 and 1990 (Table 2). Details on legislation for south and Central American coastal nations were not readily available and it may be that legislation against TBT is limited (as with many Asian nations). However, Panama (a flag of convenience nation) did state in 2003 that they would ratify the International Maritime Organisation proposed global ban (International Marine News, 2003) and Brazil has agreed to comply, subject to ratification, as have some Caribbean Island nations (Appendix 1).

A 1995 review listed 88 Marine Protected Areas in the North West Atlantic: 46 for Canada and 42 for the USA (see Mondor *et al*., 1995 for details of sites). There are varying levels of MPA status for both Canada and the USA, however, their vulnerability under MARPOL was not given. Both Canada (see: http://www.dfo-mpo.gc.ca/canwaters-eauxcan/infocentre/legislation-lois/policies/mpa-policy/system_e.asp) and the USA (see: http://www3.mpa.gov/exploreinv/status.aspx) have Government programs in place to manage and identify MPAs; the United States is aiming to have between 1500-2000 continental and oceanic sites identified in 2006. The Caribbean Region was subject to an MPA review (Stanley, 1995). Shipping pollution was listed as a significant threat to most Caribbean islands and continental states although, for example, with Panama, Mexico, Nicaragua and Colombia there was limited information on coastal environments. International efforts are being made monitor the state of marine habitats e.g. through UNEPs Caribbean Environment Program and the Caribbean Conservation association. Previous work in the Caribbean has shown very high levels (500 ngl^{-1}) of TBT in the British and US Virgin Islands (Desrosiers, 1994) and has flagged the requirement for further research in the region, as the potential for contamination may be high. Whilst data on MPAs in Venezuela were limited, the country reported to the Marine Environmental Protection Committee (MEPC, 2005) that it proposed to comply with relevant international standards with a view to reducing TBT and other pollutant inputs.

Under the 1995 World Bank and The World Conservation Union (IUCN) project to re-examine Marine Protected Areas, areas off the east and west coasts of South America were reviewed by Diegues *et al*., (1995) and Hurtado (1995) respectively. There were four protected areas listed for Argentina, but by 2001 coastal protected areas had risen to 35

(Bordino, 2001) although it was felt that there was little or no commitment to effective management and that a Governmental attitude change was needed. Some 40% of Argentineans lived in the region of Buenos Aires (Bordino, 2001) and it is likely that the majority of TBT pollution is concentrated in this region, requiring further investigation. The WWF is running the Argentina Marine Program, due for completion in 2008. This aims to form further federal MPAs and to create and improve management of coastal ecosystems (WWF, 2006[a]). In Brazil, there are 15 listed MPAs (Diegues *et al*., 1995), but in Uruguay none have been readily identified to update the legal or MARPOL status of the coastal environment.

The South America west coast is dominated by Chile, with the remainder being divided between Peru, Ecuador and Colombia; a review of MPAs in 1995 did not identify any regions in these nations MARPOL (Hurtado, 1995). Chile had 20 coastal protected areas although none of these included the sub tidal or intertidal (where TBT pollution in biota may be relatively easily detected). However, in 2003 Chile designated its first MPA 180 km southwest of Punta Arenas (a major port) and is the first of three to be created by the Chilean National Environmental Commission which is aiming to have at least 10% of the most important national ecosystems under some form of protection by 2006 (WWF, 2003; IUCN, 2006). In the 1995 review (Hurtado, 1995), Peru had no MPAs, but one has subsequently been created (Paracas National Reserve). No data were available for the impacts of TBT on coastal communities although ironically Peru is one of the world's largest producers of tin (ATSDR, 2005). Information on MPAs and other relevant coastal legislation for the Pacific coast of other South and Central American countries is limited with Hurtado (1995) identifying that between Ecuador to Guatemala MPAs were limited with only Costa Rica having several in place. No significant references were made regarding TBT pollution in any of these nation's protected areas although a section of one study had investigated TBT pollution following a shipwreck in the Galapagos (Marshall and Edgar, 2003); no impact was found.

TBT pollution in the Atlantic Gulf coast of Mexico has been investigated (e.g. Maruya *et al*., 1997; Fisher *et al*., 1999*)*, but there is limited published research for the Pacific coast of Mexico. MaciasCarranza *et al*., (1997) found TBT concentrations of 33-1021 ng/g^{-1} in sediments and 66-469 ngl^{-1} in water samples from Ensenada Harbour on the Baja peninsula. These levels were relatively high and it was noted that TBT and its degradation species were present, indicating an on-going supply of organotin pollution. The nearest protected area is the Loreto Bay National Park which is on the eastern coast of the Baja peninsula (Saenz-Arroyo *et al*., 2005). TBT pollution has formerly been a major problem on the North American Pacific coast, but in many areas TBT levels and imposex have declined (e.g. San Diego, Valkirs *et al*., 1991; Vancouver, Horiguchi *et al*., 2004) or only organotin degradation species remain in significant quantities (e.g. Santa Monica, Venkatesa *et al*., 1998). Persistent sediment concentrations may still be a cause for concern, locally (e.g. San Francisco, Pereira *et al*., 1999), however, the continued impact of TBT on North American coastal ecosystems and sensitive areas appears to be diminishing and the USA has agreed to abide by the full TBT ban once it is ratified (Appendix 1).

3.4 Arabian Gulf

The Arabian Gulf has seen limited research in to TBT pollution although work by Hasan and Juma (1992) reported high levels of TBT in sediments (128-1930 ng/g^{-1}) measured at the BAPCO terminal in Bahrain and at Hilf in Oman. More recently de Mora *et al*., (2003) investigated TBT pollution in both sediments and biota throughout the Arabian Gulf with some focus on the Gulf of Oman. TBT levels in sediment showed a maximum of $60ng/g^{-1}$, which was a considerable reduction on previous levels. De Mora *et al*., (2003) commented that "*the sharp decrease* [in TBT levels] *may reflect changing TBT usage these past years, particularly with respect to Japanese vessels*". Dry weight of organotin in muscle tissue of groupers throughout the Gulf ranged between 1.2-30 ng/g^{-1}, which is comparable with previous work in the region (Watanebe *et al*., 1998; *op cit* de Mora *et al*., 2003), but low when considered against earlier work in the Mediterranean (Kannan *et al*., 1996) and the North Sea (Kannan and Falandysz, 1998). TBT concentrations in bivalves were markedly higher than in the fish, and at Hilf in Oman levels in bivalves reflected those in sediments thus indicating recent TBT contamination at the site (de Mora *et al*., 2003). However, overall the concentrations of TBT at the survey areas in the Gulf were concluded to be low by global standards. The work by de Mora *et al*., (2003) appeared to be the most recently available published research on TBT pollution in the Arabian Gulf and despite the levels of shipping traversing the Suez Canal and operating in the Gulf, it appears the organotin pollution is minimal at the sites assessed. Five concurrent organotin surveys were reported as planned in the Gulf for 2005 (IAEA, 2005).

The Gulf of Oman, Red Sea and Gulf of Aden were all registered as Special Areas under Annex I and V of the MARPOL treaty (Chiffings, 1995). Several nations in the region had MPAs in place at that time, although for some there was limited information (e.g. Sudan, Eritrea etc). No evidence of recent research into TBT impacts upon selected MPAs or protected areas was apparent. There have been suggestions that the Arabian Gulf could become a marine sanctuary (Cava *et al*., 1993), however the latest status of MPAs in the Gulf (and elsewhere) can be checked on-line at http://www.mpaglobal.org/index.php?action=search.

3.5 Asia and India

During the last few years, monitoring of organotin contamination has been enforced in developing Asian countries (Sudaryanto *et al*., 2002). Work on TBT pollution in the Asian Pacific region, as a whole, currently appears to be more intensive than in Western Europe and the USA, where the phenomenon has been studied considerably. However, work in some Asian areas is still limited. TBT contamination in Asia and India has been of increasing concern since early work (Ellis and Patisina, 1990) showed that organotin pollution was a marked problem in gastropods from Indonesia, Malaysia and Singapore. Since that time, and unlike the African continent, there has been a wealth of research into TBT impacts upon many Asian coastal regions (where TBT is often used without legislation, despite impacts on mariculture (Bech, 2002[a]). It is possible that even though many countries may sign up to the proposed IMO TBT ban, non-signatory nations in the developing world (including the far east) may continue to develop, produce and use TBT for some years to come (Kwok and Leung, 2005).

A general overview of the situation is given here for those Pacific Rim nations where TBT research findings are readily available.

Korea Besides the classic imposex biomarker of TBT pollution in gastropods, the sensitivity of other organisms to TBT has been demonstrated in recent Asian studies. Shim *et al*., (2005) investigated levels of TBT in starfish and bivalves from the Korean coast. They found that uptake of organotin species in the starfish was correlated to levels in their prey, indicating that bioaccumulation pathways were significant and that starfish may be useful biomonitors for organotins in the region. The research did not indicate if the bioaccumulation (of triphenyltin) raised mortality in the *Asteria pectinifera* and *Asteria amuresnsis*; however, starfish are important keystone species (Paine, 1974) and pollution impacts on these organisms could have (as yet unconfirmed) community impacts. The work by Shim *et al*., (2005) highlights that TBT pollution remains a threat along Korean coastlines. Earlier work (Hong, *et al*., 2002) found that TBT concentrations in mussels (*Mytilus edulis*) from close to harbour areas were among the highest recorded at 49-2500 ng/g^{-1}. Hong *et al*., (2002) called for a complete ban or restriction of the use of TBT around Korea. In relation to MPAs in Korea, however, these were presumed to be given little importance and "*their real value for conservation of marine biodiversity is difficult to assess*" (Simard, 1995), however more recent studies may provide evidence for better management.

Indonesia In Indonesia, significant impacts of organotins on marine communities have been established for a number of years (e.g. Evans *et al*., 1995). Subsequently, port growth has been substantial (Soegiarto and Stel, 1998) and research has shown further widespread organotin contamination of coastal water mussels (*Perna viridis*) (Sudaryanto *et al*., 2002). However, recently, direct correlations between tissue burdens and total water borne tin levels have proved elusive (unlike observations in Malaysia), perhaps because of the relatively low levels of TBT in Indonesian coastal waters (Sudaryanto *et al*., 2004, 2005): nevertheless, it was recommended that organotins in the busy ports of Jakarta and Surabaya be investigated further. Indonesia has the greatest marine biodiversity in Southeast Asia and has the largest reserves of mangroves and corals in the region. Protected and endangered organisms include whales and dolphins, dugongs, turtles and molluscs. The national Government are active in marine conservation administration and several NGOs are operating in this area. However, clear information on the status of the Indonesian MPAs is scare (UP-MSI *et al*., 2002). Shipping is listed as a significant threat to the marine ecosystem of the region though this may be more significant around Java, where the majority of the population reside.

Malaysia "Mussel Watch" has been set up in developing Asian nations to monitor TBT levels in the tissues of *Perna viridis* (Sudaryanto *et al*., 2002). Using this strategy high TBT levels were recorded in aquaculture areas in Malaysia. From further work in Malaysia, Sudaryanto *et al*., (2004) noted that high organotin levels were found in all of the *P. viridis* samples taken, and that concentrations in fish tissues ranged between 3.6-210 ng/g - a possible concern for human health. Although the degradation compounds dibutyl- and monobutyl-tin were found in environmental samples along the coastline, the most common tin species was TBT "*suggesting recent input of TBT to the Malaysian marine environment*" (Sudaryanto *et al*., 2004). It was concluded that Malaysian commercial vessel traffic had vastly increased, resulting in TBT and tin species levels being amongst the highest recorded in any developing Asian country, with hot-spots near to harbours and general pollution by tin in all of the sites sampled. Sudaryanto *et al*., (2004) also stated that, in the absence of regulation, use of organotins "*posed a serious pollution threat in the future*". Coral reefs, barrier reefs, sea-grass and mangroves are common marine habitats in Malaysia. Species of conservation concern are similar to those in Indonesia. There are several MPAs in Malaysia, and the conservation and protection system is well developed (UP-MSI *et al*., 2002). No direct reference was made to potential impacts upon these from shipping, however surveys showed that the "*most serious and common threat* [to marine areas] *is coastal development*" (UP-MSI *et al*., 2002)

Taiwan In Taiwan, coastal waters have been polluted by organotin concentrations as high as 560 ng l^{-1} (Lee, 2001). Hung *et al*., (2001) have recorded high imposex levels in gastropods on the Taiwanese coast and high TBT levels in oyster populations (*Crassostrea gigas*). Pollutant contaminated seafood has been reported as a cause of ill effects in humans (Han *et al*., 2002) notably through tainted oysters (Chien *et al*., 2002), though the links with organotins were unknown. Recently TBT levels in fish used for human consumption were investigated; levels were only significant for pelagic fish from the Wuchi fishing harbour (Lee *et al*., 2005). However, the economy in Taiwan, as in many far eastern nations, appears to be growing, and in the absence of stringent regulation, TBT pollution may require regular monitoring to ensure risks do not become unacceptable, either to humans or marine conservation areas. Taiwan was previously listed as having two MPAs, Kenting National Park and Tansui Mangrove Forest (Simard, 1995), habitats include coral reefs and mangroves (and presumably TBT-sensitive gastropods which inhabit mangroves). Further research would be useful to assess impact, if undertaken with caution; the protected mangrove area in Taiwan is not large (76.5 Ha) and may not support high levels of disturbance.

Thailand TBT impacts in Thai waters from both commercial shipping and recreational vessels are well established with, for example, Swennen *et al*., (1997) reporting up to 100% imposex in sublittoral gastropods from the Gulf of Thailand and straits of Malacca (related to distance from main shipping routes). In addition the boom in tourism and recreational vessel use in Thailand has resulted in greater boating traffic around marinas and using a mangrove dwelling gastropod (*Chicoreus capucinus*) Bech (2002[a]) found that imposex developed significantly at a site of marina development on Phuket Island. For comparison, another gastropod *Thais distinguenda* was used to check imposex incidence at isolated bays on Raja and Phi Phi Islands (south east of Phuket). Event though there were no marina facilities, TBT from visiting craft caused imposex in the gastropod and highlighted how TBT from casual visiting yachts in isolated areas can affect sensitive species. Further studies by Bech (2002[b]) "*suggest that TBT contamination is worsening, against global trends, because regulations prohibiting the use of TBT-based paints, do not exist in Thailand*". Thus it seems likely that TBT levels impacting the marine biota of Thailand, affected by commercial *and* recreational shipping, will worsen unless legislation is adopted. There are 21 MPAs in Thailand of which three are globally significant. In a recent review of their status, TBT was not mentioned as a specific threat, though their management was described as weak (UP-MSI *et al*., 2002). With the likelihood of continued and worsening TBT use in the region and limited management of MPAs, greater monitoring and impact assessment of TBT from ports and marinas should be considered.

Vietnam In Vietnam, few studies have been conducted to assess the impact of organotins. Sediment-bound TBT has impacted upon clam (*Meretrix* spp) populations, which are part of the local human diet (Midorikawa *et al*., 2004). Further information on TBT pollution in Vietnam was not readily available, however the "*ongoing economic liberalisation*" (Midorikawa *et al*., 2004) of Vietnam has led to increasing trade with other countries and thus organotin impacts may require further assessment. Vietnam has two MPAs (Cát Bá Islands and Halong Bay National Park and the Côn Dảo Islands National Park). Both are designated for fringing coral reefs and the Côn Dảo Islands National Park is also recognised for its mangroves (Bleakley and Wells, 1995). Ha Long Bay is a UNESCO World Heritage Site and has been subject to review and management by the Vietnamese Government. From 1998-1999 an Environmental Impact assessment was conducted for the impact of the port development at Cailan (ESSA, 2006) after requests from UNESCO, and specific reference made to the need to keep large ships away from the bay (UNESCO, 1997).

China (and Hong Kong), India and Japan Of the significant industrialised nations that require detailed consideration in this section, Japan, India and China are the most noteworthy, due to their growing economies, particularly India and China.

China China is one of the most rapidly developing economies in the world and industrial development in the region has led to increased concerns about pollution effects. However, there were few studies of organotin impacts in China until 2001, when it was recognised that TBT from antifouling paints was a serious contaminant in both marine and freshwater areas (Jiang *et al*., 2001). TBT degradation products (i.e. mono and dibutyltin) were found in most of the freshwater and marine habitats surveyed and TBT levels were above the toxic thresholds for aquatic organisms. Concerns were raised, both for the ecosystems themselves and for humans who utilised the water resources, although it was highlighted that the long-term chronic effects were unknown (Jiang *et al*., 2001). Since then, further research has considered the impact of TBT on the clam *Meretrix meretrix* which is used for human consumption in China (Wang *et al*., 2005), as has also been the case in Vietnam (Midorikawa *et al*., 2004). Hormonal changes in the clam were detected and reproduction potential reduced, which may have long-term commercial implications. Subsequent research on Chinese coastal waters has found "*wide occurrence and serious pollution of BTs* [butyltins] *in seafood*" and suggested this may be a danger to human consumers (Yang *et al*., 2006). Impacts of diffuse TBT pollution from shipping may be set to increase in China as a whole, as the levels of container transport for China's global trade have increased from 2 million TEUs (standard container capacity measure of 20-foot equivalent units) in 2001 to around 6 million TEUs in 2005 (Merrill Lynch, 2006).

China (Hong Kong) In contrast to the rest of China, TBT pollution in Hong Kong has been studied for some years (Morton and Blackmore, 2001). Morton and Blackmore (2001) used the marine environment in Hong Kong Harbour as analogous to what may have already occurred in South China seas in terms of marine pollution. High TBT levels in Hong Kong Harbour were reported as long ago as 1991 (Lau, 1991) where TBT was detected in clams, sediments and marinas. Subsequently levels were reported as 500 ngl^{-1} in some marina sediments and typhoon shelters (Ko *et al*., 1995) although high TBT levels have shown considerable spatial variance (EPD, 2002). Morton and Blackmore (2001) reasoned that a combination of the high tonnage of commercial craft and illegal usage of TBT on small craft accounted for high TBT levels in Hong Kong Harbour. As China opens up to increasing global trade, levels of TBT in biota and sediments should be monitored, particularly if the ban is not fully adopted or, as seen in small craft (Morton and Blackmore, 2001) is largely ignored. China is increasing its focus on MPAs and the "First Asia Pacific Coral Reef Symposium" is planned for June 2006 in Hong Kong. The State Oceanic Administration (SOA) is responsible for MPAs in China and in 1993 there were 41, covering 4,553 square kilometres. In 1995 Simard reported that the SOA had been very active, but that it was too early to evaluate success.

India In the Mussel Watch program already identified here, data from India between 1994 and 1997 generally suggested less TBT contamination than in Thailand or the Philippines. However, it was highlighted that economic growth in the region may result in the increased use of TBT (Tanabe *et al*., 2000). TBT has been found in harbour sediments in India, with west coast levels higher than east coast (Rahendran *et al*., 2001). Bhosle *et al*., (2004) commented that India has not implemented TBT legislation (see also Garg and Bhosle, 2005) and in their research found sediment samples containing between 5 and 2853 ng/g of TBT. The degradation species of organotin were not as abundant as TBT, thus it was concluded that TBT inputs were fresh. Recent work has shown the TBT levels in sediments of Kochi Harbour were 16 - 16,816 ng/g and in Mumbai were 4.5 - 1193 ng/g. These include concentrations, which are high enough to induce toxicological effects in sensitive species (Bhosle *et al*., 2006). Garg and Bhosle (2005) measured TBT contamination in oysters *Saccostrea cucculata* in west Indian Harbours and suggested a link between rapid economic growth and unregulated TBT pollution. Links between marine pollution and economic growth in the region have also been made previously by Glasby and Roonwal, (1995), thus, where feasible, TBT inputs in Indian coastal regions may require further monitoring. Indian MPAs cover 1,593 ha (Earth Trends, 2003), and growth in India, and the associated increase in shipping has been identified as potentially raising the threat of TBT pollution, (Tanabe *et al*., 2000) and, since "*India has important examples of all the main ecosystems found in the region*" (Wells *et al*., 1995), the registration of sensitive areas may require review.

Japan In contrast with the majority of countries in the Asia-Pacific region, Japan showed a pioneering approach to combating organotin pollution - from a very early stage in the debate over detrimental effects of these compounds. After early monitoring confirmed contamination (e.g. Tanabe, 1988), legislation banning all organotins (TBT and TPT) on coastal vessels was put in place in 1990. However, the legislation has seen varying levels of success with TBT pollution still apparent in areas of industrial shipping (Murai *et al*., 2005). Open coastal water TBT pollution was investigated between 1994 and 1997 by collecting Caprella shrimps from seaweed (*Sargassum* spp) clumps around north Japan, the Sea of Japan, Tokyo Bay and adjacent western areas. Takeuchi *et al*., (2004) found that TBT was detected in all samples, even in unpopulated areas where industrial activity was low. It was also concluded that there were hot-spots of TBT pollution despite the legislation, which was "*not effective enough*". TBT levels in the giant edible abalone *Haliotis madaka* were studied between 1998 to 1999 in both control (unimpacted) and treated (organotin affected) populations (Horiguchi *et al*., 2005). Nineteen percent of the females from the polluted site had evidence of hormonal disruption, with male sex organs apparent and a population decline in this and other members of the abalone family in the area. The rearing of oysters for both human consumption and pearl culturing has also been affected in some Japanese coastal areas (Ramaswamy *et al*., 2004). TBT was painted on fishing equipment and sediment levels of TBT were found up to 2500 ng/g. Ramaswamy *et al*., (2004) suggested the need for further regulatory action and continued monitoring. As a departure from threats to molluscs, recent work in Japan has included impacts in fish - for example the possibility of maternal transfer of TBT in the perch species *Ditrema temmincki* (Ohji *at al*., 2006). This research showed a cumulative impact of TBT during transfer to juveniles from adult females. It was found that the threat of bioaccumulation in the young was heightened by a reduced ability to eliminate TBT.

Japan was one of the first nations to call for a total ban on the use of organotins and has imposed stringent legislation against these compounds. However, their continued presence in the environment suggests that the legislation may not be entirely effective (Takeuchi *et al*., 2004) and that continued illegal use is a problem (Ramaswamy *et al*., 2004). Murai *et al*., (2005) commented that TBT pollution had implications for community structure of marine organisms in the areas investigated and that the productivity of higher trophic fishes may have been affected.

Japan has National Parks and a Fisheries Resources Protection System, however, these were described as inadequate and the high population density and industrial heritage of Japan may not be conducive to successful MPAs in the region (Simard, 1995). There are no direct references to the impact of marine pollution on MPAs or any of the protection systems in Japan, however in East Asia and Micronesia a current reassessment of MPAs is taking place with particular reference to coral reef habitats (ICRI, 2005). This is being coordinated through Japan.

3.6 Australasia

This section provides an overview of recent TBT research relevant to the main landmasses of Australia and New Zealand. However, TBT pollution has been investigated in South Pacific Islands where some significant results have been obtained for Fiji and Guam (see: Stewart and de Mora, 1992; Maata and Koshy, 2001; Denton *et al*., 2005) and further research has been recommended (Denton *et al*., 2005).

Both Australia and New Zealand placed restrictions against the use of TBT in 1989 (Table 1) with New Zealand further restricting the use of specific TBT paint types in 1993 and Australia banning the reapplication of TBT in Australian docks on any vessel from 2006 (Environment Australia, 2003). Bioindicators have been used to demonstrate the effectiveness of legislation in reducing TBT contamination. For example, Batley *et al*., (1992) showed improvements in the commercial oyster *Saccostrea commercialis* population in Sydney Harbour, following the partial banning of TBT, whilst Rees *et al.,* (2001) reported similar success in the recovery of *Thais orbita* in Port Philip Bay on the New South Wales coast. It was found that, on a regional scale, *T. orbita* had largely recovered from TBT and that populations and imposex indices were improving. There were sites, however, where both recreational and commercial boats were used and imposex levels had not improved. Reitsema *et al*., (2003) surveyed 16 sites around Perth in Western Australia metropolitan area and reported a significant reduction in imposex in *T. orbita* at 11 sites, relative to data from 1991. However, at locations near commercial docks the prevalence of imposex in *T. orbita* was 100% with almost one-third of females permanently sterile (Reitsema *et al*., 2003). Thus, recovery is not always clear, and continued monitoring of imposex in gastropod bioindicators provides an important resource highlighting the mixed effectiveness of legislation. Supplementing this approach, Gibson and Wilson (2003) used a combination of *T. orbita* transplants and local populations to study TBT-induced imposex in Sydney harbour in further detail. It was found that TBT levels remained sufficiently high to induce imposex quickly, but that the overall severity had declined in recent years, suggesting a gradual improvement in TBT concentrations. However, Gibson and Wilson (2003) found hotspots of contamination in Sydney harbour, which is consistent with patterns seen at other busy ports, world-wide. Smith (1996) used similar gastropod imposex indices to discern the effectiveness of TBT legislation in New Zealand. As in other countries with partial legislation, it was found that TBT levels had declined in the vicinity of recreational harbours, but where commercial shipping was prevalent, TBT pollution was slow to abate. TBT residues in sediments were particularly persistent, with concentrations at some sites decreasing little over a 15 years period (de Mora *et al*., 1995). Other work in both New Zealand and Australia has focussed on the continued legacy of TBT in sediments.

The levels of sediment bound TBT were investigated in New Zealand a decade ago by Roberts and Forrest (1999), though on this occasion, the impact of dredge spoil on sensitive areas in Tasman Bay was found to be negligible (contrary to other sites e.g. Santos *et al*., 2004) due to the dynamic dispersing nature of the area and enhanced mixing of sediments. Burton *et al*., (2005) investigated TBT in sediments at a commercial marina (south east Queens land, Australia) and found that substantial degradation had occurred highlighting the potential efficacy of partial legislation, even on sedimentary sinks. However, considering that TBT was banned on recreational craft some 16 year ago in Australia, the fact that it can still be widely detected in sediments highlights the need for appropriate risk assessments and potentially greater control, particularly when sediments are disturbed, either naturally or deliberately.

In summary, despite the restrictions on TBT in the late 1980s to early 1990s, Australia and New Zealand (along with many other countries) may still have a legacy of TBT in sediments, particularly in fines in sheltered embayments and estuaries. Disposal of such material after dredging operations may require careful consideration. Furthermore, some hotspots still have high waterborne concentrations of TBT, particularly where commercial shipping is prevalent.

The potential for TBT impacts on sensitive and protected areas in Australasia have been recognised for some years (e.g. Batley, 1995). The major ports in Australia are Brisbane, Sydney, Melbourne, Adelaide and Freemantle with smaller regional ports also handling industrial shipping dotted in between. Marine Protected Areas ring Australia's coast, (see: http://www.deh.gov.au/coasts/mpa/) including the Great Barrier Reef, which may have been impacted by elevated TBT levels (see: Negri *et al*., 2002; Jones *et al*., 2005). The demonstrated effects on the Great Barrier Reef have so far been very specific and localized (due to a ship grounding), however, considering the importance of the site, the continued monitoring of TBT influences on this and Australia's other marine ecosystems is of paramount importance. The Government of Australia and its conservation organisations are highly aware of their marine heritage and are working towards the total ban on TBT.

The coastline of New Zealand is not developed excessively and there are only four major ports serving the nation (Auckland, Dunedin, Lyttleton, Wellington). There are several Marine Reserves associated with Auckland, two sites close to Wellington and one at Christchurch. No sites are listed adjacent or close to Dunedin (see: http://www.doc.govt.nz/Conservation/Marine-and-Coastal/Marine-Reserves/index.asp). Survey work at Wellington Harbour (Smith, 1996) found biologically significant levels of TBT and high imposex levels have been determined in indicator species close to Auckland (Stewart *et al*., 1992); however there have been few recent reports on TBT pollution in these sensitive areas.

3.7 Europe, Baltic and Mediterranean

Whilst the potential for TBT toxicity to non-target species was first suggested in North America (Thomas, 1969; *op cit* Spence, 1989), the first legislation against the compound was put in place in France (Alzieu *et al*., 1986) and the clear link between TBT and imposex was established in the UK (Bryan *et al*., 1986). Since partial bans against TBT were put in place in Europe (Table 1), water and sediment concentrations around some recreational harbours have declined appreciably (e.g. Mediterranean, Diez *et al*., 2002) and in such situations imposex in gastropod populations and body burdens of tin in bivalves have decreased correspondingly (e.g. Ireland, Minchin, 2003; Spain, Gómez-Ariza *et al*., 2000; UK, Harding *et al*., 1997; Hawkins *et al*., 2002; North Sea, Birchenough *et al*., 2002). This has affected the recovery of benthic fauna. For example, since the partial ban on TBT (for vessels < 25 m), the epifauna in the Crouch estuary (a south east UK yachting area) has improved between 1987 and 1997, with an increase of sedimentary taxa and return of species such as *Ostrea edulis* (Rees *et al*., 1999; Waldock *et al*., 1999). However, throughout Europe there have been concerns about continued hot-spots of contamination (e.g. Germany, Nehring, 2000; Mediterranean, Diez *et al*., 2002; UK, Morgan *et al*., 1998; Hawkins *et al*., 2002). This applies particularly to contaminated sediments (e.g. Baltic, Bisselli *et al*., 2000; Mediterranean, Diez *et al*., 2002; Sweden, Brack, 2002; UK Thomas *et al*., 2000, 2001; Harino *et al*., 2005[a]). The issues associated with the disposal of TBT-polluted dredged sediments have been articulated frequently (e.g. Weichsel *et al*., 1998; Svavarsson *et al*., 2001; Santos *et al*., 2004).

The problems of imposex and tissue burdens of tin are ongoing in Europe, at least in places. For example surveys in the Venice Lagoon found high levels of imposex and body burdens of tin (102±17 to 432±27 ng Sn g^{-1}) in the gastropod *H. trunculus* (Pellizzato *et al*., 2004). Describing TBT contamination along the coast of Corsica, including in protected areas, Michel *et al*., (2001) felt there was "*little hope for further improvement unless the phase-out under consideration by the IMO is applied*". Terlizzi *et al*., (2004) found a similar TBT problem in populations of *H. trunculus* from remote sites on the Italian coast, which were all within MPAs. Conversely, at other sites there has been encouraging evidence of recovery in gastropod (*N. lapillus*) populations, which have recolonised sites where they were formerly extinct due to TBT; amongst other locations this has even been found on the formerly ubiquitously polluted UK south coast (e.g. Herbert *et al*., 2000; Colson and Hughes, 2004). These latter findings suggest a general improvement throughout the UK south coast region. However there are still sites such as parts of Southampton Water where sediment and waterborne TBT levels are high, (Thomas *et al*., 2000, 2001; Galloway *et al*., 2004). Trial reintroductions of *Nucella lapillus* populations within Southampton Water showed that severe imposex developed within six months (Bray, 2005). This was despite the legislation restricting TBT use on the numerous recreational vessels in the area, thus implicating TBT from commercial shipping. In addition in the case of Plymouth Sound and its estuaries, TBT levels are sufficient to delay the recovery of impacted *Nucella* populations (Hawkins *et al*., 2002; Bray 2005).

Atlantic Coast of Europe

Despite its relative exposure to strong currents and wave action, the Atlantic coast and its sheltered waterways on mainland Europe are still suffering from organotin pollution. The recognition of TBT pollution in Arcachon Bay, south-western France, which adversely affected oyster (*Crassostrea gigas*) populations, created the impetus which led to early organotin legislation (Alzieu *et al*., 1986; Alzieu, 1991). Twenty years after the ban in France (Table 1) Devier *et al*., (2005) reported work using several biomonitoring species and sediment sampling to establish TBT levels in Arcachon Bay. Despite the ban in 1985, TBT levels were at a background level of 30 ngl^{-1} and tin levels in Arcachon Harbour were at 1500-2000 ng Sn g^{-1}. This serves to highlight the persistence of organotins in sediments and that this TBT 'reservoir' may require consideration for some years to come.

Below France, in the Rias of northern Spain, TBT pollution was widespread in the region, with particularly high levels reported close to centres of shipping (Ruiz *et al*., 1998). By transplanting healthy gastropods (*N. lapillus*) to sites within the Spanish Galician Rias it was possible to demonstrate endocrine disruption in the form of imposex (Quintela *et al*., 2000). Recent biomonitoring work by Ruiz *et al*., (2005) describes continued organotin pollution in the Galician Rias. The authors also comment that "*the previous partial* [TBT] *ban in European waters appears to have taken much more rapid effect in the UK than in other countries such as France, Portugal and Spain*". Furthermore Prego and Cobelo-Garcia (2004) reported that not enough work had been undertaken in the region to investigate the concentrations of TBT and that much of the work focussed on areas of high socio-economic activity. Thus it may be necessary to make further investigations of the Galician Rias to consider sites more distant from industrial shipping.

Further down the Atlantic coast of Europe, Portugal has been the focus of both biological and sediment-based monitoring of TBT levels. As early as 1988 imposex in *N. lapillus* was reported by Peña *et al*., (1988) and subsequent work has established that the bioindicator species is affected along much of the length of the Portuguese coast (Santos *et al*., 2000; Galante-Oliveira *et al.,* 2006); these surveys are valuable baselines to monitor the efficacy of legislation against TBT. Subsequently Santos *et al*., (2002) has indicated that TBT pollution appeared to have increased from initial survey results, implying that the partial legislation put in place in Portugal in 1993 was ineffective. This observation was echoed in work by Barroso and Moreira (2002) in which imposex and body burdens of tin in *N. lapillus* and *Nassarius reticulatus* (now *Hinia reticulata*) were found to have increased over earlier values (e.g. Peña *et al*., 1988) in some areas. Barroso and Moreira (2002) felt that the apparent inefficacy of Portuguese legislation highlighted the need for success in the IMOs proposed total ban of organotins. Since then Diez *et al*., (2005) reinvestigated TBT pollution at 46 sites along the Portuguese coast and found that TBT levels were "*remarkably low*" in comparison with other European countries and felt that the level did not pose a risk to marine populations. However, in common with other locations around Europe, hot-spots were present – for example in the Algarve region of southern Portugal (Santos *et al*., 2002). This has also been found by Vasconcelos *et al*., (2006) who reported highly affected (greater than 90%) masculinised female *H. trunculus* in the Ria Formosa, Algarve. Hot-spots are also often associated with commercial ports, which require consistent maintenance dredging to ensure vessels can navigate freely (Galante-Oliveira *et al.,* 2006). This can lead to the secondary impact of TBT being released from sediments during dredging and spoil-disposal operations. Santos *et al*., (2004) considered the potential impacts of dredged material from the Portuguese port of Oporto on offshore populations of *H. reticulata* and showed that imposex levels were inversely related to distance from the dredge spoil dump site. The impact was lower, overall, than that in *H. reticulata* from moderately contaminated inshore sites, however the results highlight that care is needed when disposing of dredge spoil, particularly where there is a risk it may be re-mobilised by hydrodynamic forces into sensitive or protected areas. In a further study using *Hinia* as a biomonitor along the NW Portuguese continental shelf, the effects of TBT were shown to extend considerable distances offshore of sources in the Ria de Aveiro (Rato *et al*., in press).

Baltic

The Baltic is designated as a special area under MARPOL 73/78 and amongst other natural features, is characterised by a very low flushing rate (MEPC, 2001) leading to long retention times for pollutants. In a 1998 report to the WWF Weichsel *et al*., (1998) describe the presence of high TBT levels in sediments along parts of the German Baltic coast and indicate that these 'hot-spots' were locally associated with marinas. Widespread masculinisation and hormonal disruption in gastropods was reported (e.g. in *Hydrobia ulvae* and *Littorina littorea*) and there was "*concern for future practice in dredging and disposal of TBT-contaminated sediments*". Subsequently, more work in the Baltic Sea has registered high levels of sediment-bound TBT (Biselli *et al*., 2000). Germany placed restrictions on TBT use in 1989/1990 (Table 1), however at marinas in the German section of the Baltic, TBT was found in sediments at levels of 570-17,000 ng/g^{-1}. For comparison, in the North Sea, with greater water exchange, TBT in sediment ranged between 80-720 ng/g^{-1} (Biselli *et al*., 2000).

Work on TBT-sediment related affects on biota in the Baltic was carried out by Falandysz *et al*., (2002). They investigated TBT impacts on the three spined stickleback (*Gasterosteus aculleatus*) from the Gulf of Gdańsk, Poland. Amongst other locations investigated it was found that sediments in the marinas close to the city of Gdynia contained tin levels between 130-20,000 ng/g^{-1} and that the body burden of tin in stickleback samples ranged between 1500-3100 ng/g^{-1} (wet weight). It was concluded that in Poland there had been continuous use of TBT on recreational craft less than 25 m long (Falandysz *et al*., 2002). These findings have implications for higher organisms and studies on the impact of TBT on marine mammals in the Polish region of the Baltic have been undertaken. High levels of TBT were found in the livers of stranded or netted marine mammals throughout the Gulf of Gdańsk, with tin levels ranging from 43.9-7698 ng/g^{-1} (dry weight) (Ciesielski *et al*., 2004); it was concluded that there was a significant degree of TBT pollution along the coast of Poland. Subsequent work on harbour porpoises has led to the recommendation that "*organotin compounds…should be integrated when assessing the risks of contaminants on the health and viability of harbour porpoises in the…Baltic Sea*" (Strand *et al*., 2005).

The entry into the EU by former eastern block nations will require their compliance with relevant environmental legislation, including that pertaining to TBT. Thus, the impacts of TBT pollution from nations such as Poland are expected to decline eventually; however the relatively high levels of TBT contamination present in some sediments and biota in the region indicate that management of dredged material will require careful consideration in the interim period.

Mediterranean

Similarly to the Baltic the Mediterranean has a low flushing rate and high residence time for water masses (e.g. Ferreira *et al*., 2005). When compared to the North African coast, the European north western Mediterranean coastal region has higher pollution levels possibly due to greater industrial development (Gabrielides, 2000). In the western Mediterranean Diez *et al*., (2002) investigated organotins in sediments at 38 locations around Spain. TBT was widespread throughout the whole area, which indicated that recent inputs of organotins had occurred in the region and that these were associated with both commercial and recreational/fishing harbours. However, levels in commercial harbour sediments average at 5000 ng/g^{-1} and in fishing/recreational harbours at 1000 ng/g^{-1}. It was concluded that levels were comparable with those in other developed nations (e.g. see Thomas *et al*., 2000, 2001) where regulations were in effect (*cf*. Polish Baltic sediment levels).

Working on imposex in *H. trunculus* populations on Malta in the central Mediterranean, Evans *et al*., (2000[b]) concluded that the unacceptable level of TBT pollution on the island justified a total ban on TBT, adding the caveat that biomonitors had limitations in the empirical identification of imposex causation. Nevertheless, there has been considerable use of *H. trunculus* in assessing TBT impacts in the Mediterranean region. Recently, as previously discussed, very high imposex levels in *H. trunculus* were found in populations at the Lagoon of Venice (Pellizzato *et al*., 2004) and further work has suggested a synergistic role for polychlorinated biphenyls and pesticides, alongside organotins, in producing effects (Maran *et al*., 2006).

Whilst many of the northern Mediterranean nations are bound by legislation to restrict TBT use, the low-energy nature of the sea may mean that levels will persist for some time. As suggested by Gabrielides, (2000), TBT pollution in the Western Mediterranean may be associated with higher industrial capacity in the region, which could, in turn, result in TBT impacts in MPAs: there are already established records of pollution impacts in MPAs showing the ability of contaminants to cross conservation boundaries in the Mediterranean region (Michel *et al*., 2001; Terlizzi *et al*., 2004). There is even speculation that TBT pollution could increase over the next few years (as TBT coatings are removed from vessels, prior to the 2008 ban), before finally disappearing (e.g. Champ, 2000). Thus, there is clearly potential for continued impact from organotins in the slow-flushing Mediterranean Basin.

United Kingdom

The UK was the focus of much of the early research into the impacts of TBT on marine fauna, commencing with studies on shell-thickening in oysters *Crassostrea gigas* (Waldock and Thain, 1983). The system to quantify levels of TBT pollution by assessing imposex in neogastropods was developed by scientists at the Marine Biological Association of the United Kingdom (Bryan *et al*., 1987; Gibbs *et al*., 1987). In addition levels of organotins in sediment and their impact on sensitive species have been monitored since the 1980s (see Langston *et al*., 1990; Langston and Burt, 1991; Langston *et al*., 1994). The severity of imposex has improved in numerous UK *N. lapillus* populations in recent years, following the partial ban on TBT in 1987, allowing some populations to recover or recolonise as TBT levels decline (Colson and Hughes, 2004). For example Bray and Herbert (1998) found that *N. lapillus* had recolonised sites on the north coast of the Isle of Wight (UK south coast), where, previously, all *N. lapillus* populations had become extinct (Herbert, 1988). Subsequent work has shown that numbers of *N. lapillus* at these sites are increasing (Herbert *et al*., 2000; Bray 2005). A similar scenario was recorded by Crothers (1998) who found that a previously extinct *N. lapillus* population had recolonised Watermouth Cove, (north Devon) and that numbers were subsequently increasing (Crothers, 2003). Birchenough *et al*., (2002) also reported widespread recovery of *N. lapillus* populations at sites where extinction had occurred (Isle of Cumbrae, Scotland, north-eastern English coast, southwest English coast), attributing this to efficacy of legislation and the development of slow release paints. Interestingly, these authors suggested caution in switching to TBT alternatives for which biological impacts were unknown.

Whilst recovery is apparent throughout much of the UK, and Europe, there are locations where TBT pollution is still high. Amongst other antifouling compounds, Thomas *et al*., (2000, 2001) investigated TBT levels in Southampton Water (UK south coast). They found TBT levels at recreational marinas in the area were "*below or near the environmental quality standard (EQS) of 2 ng/l*" but that levels were much higher in some marinas and sites of commercial vessel operation. The persistence of TBT in sediments and a range of biota from dockyard-impacted sites in the Test, such as Cracknore Hard, was also previously described by Langston *et al*., (1994). Here TBT may be bound in sediments in association with organic matter and paint flakes and has an extremely long half-life, posing a continuing long-term risk to organisms such as infaunal clams (Langston *et al*., 1994; Galloway *et al*., 2004). This contrasts with the Hamble estuary (small-boat dominated) where the partial ban was initially highly effective in reducing TBT concentrations (Langston *et al*., 1994).

The long-term legacy of TBT in UK sediments subjected to commercial shipping is apparent from core samples in Tilbury Docks. A core taken in 1994 was analysed for TBT content to indicate degradation of the compound (from inputs introduced in the 1960s). By using a degradation rate constant, Scrimshaw *et al*., (2005) estimated that TBT at levels of 20-60 ng/g^{-1} in the 1994 sediment core extrapolated to values of 430-600 µg g^{-1} in the 1960s. It was concluded that TBT persists for significant periods in anaerobic sediment and that "*remobilisation may introduce bioavailable inputs of TBT to the environment which may become relatively more significant as the ban on TBT comes into effect*" (Scrimshaw *et al*., 2005). Such persistence may have implications through community food webs. Harino *et al*., (2005[b]) investigated TBT bioaccumulation in benthic biota in the Mersey Estuary and found biomagnification (accumulation of TBT in tissue relative to sediment). From the levels encountered they concluded that the dredging of TBT-rich sediments posed a problem for future disposal, which required careful management, and that baseline data for other major ports would be useful to monitor potential consequences for biota. In addition, the Mersey is a RAMSAR site and a Special Protection Area (European Wild Birds Directive). Seabirds have been shown to bioaccumulate TBT through predation on polluted prey (e.g. Guruge *et al*., 1996), thus interactions such as this may have implications for protected bird species at sites such as the Mersey estuary.

Marine Protected Areas

European states have had MPAs, or similar, in place for several years and numerous sites are covered by EU legislation (e.g. Marine Special Areas of Conservation (SAC), under the European Habitats and Species Directive (92/43/EEC); Special Protection Areas (SPA), under the European Wild Birds Directive (79/409/EEC) together with international designations such as RAMSAR (for wetlands of international importance). A review of MPAs and international equivalents was carried out by Gubbay, (1995) in which the only site designated under MARPOL was the central North Sea. Since then, European countries have nominated a host of SACs and SPAs around their coasts (see http://www.jncc.gov.uk for UK sites). A further UK example of conservation efforts that consider the potential for impacts of TBT is the UK and Local Biodiversity Action Plan (UKBAP, LBAP) for habitats and species. Amongst other threats, habitats and species sensitive to TBT are listed on a regional and national basis.

The WWF launched a project in 1999 to create a network of MPAs based on the OSPAR region (see: http://www.ospar.org/eng/html/welcome.html). The main aim is to "*promote and help establish a network of MPAs in the North-East Atlantic in order to protect a representative section of its species, habitats and ecological processes*" (WWF, 2006[b]). The impacts of TBT on protected areas have been seen in European waters (Michel *et al*., 2001; Terlizzi *et al*., 2004) and the potential for TBT bioaccumulation to higher organisms has also been observed (Guruge *et al*., 1996). Thus the synergistic approach offered by the WWF MPA project should be welcomed, as future issues with the disposal of contaminated sediment from European nations (and elsewhere) will require careful management.

3.8 Polar Regions

TBT pollution may not be seen as a factor within Polar Regions, however this has been a subject of some debate. Tributyltin pollution has previously been recorded and monitored in sub-Arctic Icelandic waters (e.g. Skarphéðinsdóttir *et al*., 1996; Svavarsson, 2000) where general improvements in imposex in *N. lapillus* populations have been recorded (as a result of earlier legislation). More recently, through gastropod monitoring, Jörunsdóttir *et al*., (2005) found further evidence of TBT reductions around Iceland's coast, but that levels were still high in the major harbours. Within the Arctic region TBT contamination has been found in Harbour Porpoises (*Phocoena phocoena*) from the west coast of Greenland (Strand *et al*., 2005) and although this was at low levels it does indicate the spread of TBT to Arctic regions.

Organotins have been recorded in cetaceans from the Antarctic region (Tanabe, 1999) and, more recently, concern has been raised after the discovery of TBT in Antarctic marine sediments (Negri *et al*., 2004). This has heightened the debate regarding the necessity of antifouling paint use on vessels in Polar latitudes. Suggestions have been made that antifouling paints are not necessary for ice breakers moving deep within Polar waters, as the action of ice on hulls scrapes off fouling organisms (New Scientist, 2004). Conversely it has been argued that the threat of alien marine organisms being transported on hulls to the ecologically sensitive Antarctic may be a greater threat than the TBT impacts, themselves (Lewis *et al*., 2004).

The nations administering Arctic waters have established several MPAs, as recorded by a review in 1995 (Bleakley and Alexander, 1995). In addition, much of the region was designated as UNESCO 'Man and Biosphere reserves'. No published records of TBT in specific conserved areas within the Arctic were found, however, the difficulty of research in this region may be restrictive. The whole of the Antarctic is designated as an MPA under IUCN rules and the continent is also covered by the Antarctic Treaty System (Dingwall, 1995). Under these designations the work by Tanabe *et al*., (1999) and Negri *et al*., (2004) shows that TBT is present in an internationally conserved area and that a legacy may be present in sediments.

3.9 Oceanic

Evans and Nicholson (2000) concluded that open oceanic TBT pollution was not present at biologically significant levels. They questioned research by Ten Hallers-Tjabbes *et al*., (1994) which demonstrated imposex in North Sea populations of the edible whelk *Buccinum undatum*, although this has been confirmed more recently by other workers (e.g. Strand and Jakobsen, 2002). Further evidence of offshore transport of TBT can be found in body burden data: for example, high levels of organotins have been measured in the tissues of diseased and stranded cetaceans (Tanabe, 2002). De Brito *et al*., (2002) collected deep sea benthic and pelagic organisms from the North Pacific off of Japan and whilst highest organotin levels were often associated with coastal areas, which receive regular, or recent inputs of organotins, they found that residues in deep sea samples were becoming "*close to reported values of estimated effect concentrations*". The authors recommended further studies on the effects of organotin pollution on deep-sea organisms and ecosystems. This suggestion may be appropriate in the light of more recent concerns at the disposal of TBT-contaminated dredge spoil (e.g. Santos *et al*., 2004). Finally, Ueno *et al*., (2004) have used skipjack tuna (*Katsuwonus pelanus*) as an effective biomonitor for organotin pollution throughout the oceans. Organotins were recorded in all of the samples taken, with particularly high levels recorded off Japan and from the offshore waters around developing Asian nations. It was concluded that there was widespread contamination of butyltins on a global basis. Whilst the open ocean has no statutory legislational control, there are protected oceanic species that have been influenced by organotin exposure: for example the IUCN Red List species beluga whale (*Delphinapterus leucas*) have been shown to accumulate butyl tins (St Louis *et al*., 2000). Clearly there is the potential for impact in oceanic species and Ueno *et al*., (2004) suggest this may be worsening.

3.10 Freshwater

Whilst not a primary aim of this work, a short consideration of TBT impacts on freshwater habitats highlights the toxicity of the compound in all aquatic environments. TBT pollution has been reviewed in landlocked nations and Austria and Switzerland have banned the compound TBT in their lakes and rivers. Becker Van-Slooten. and Tarradellas, (1994) monitored TBT impacts on the freshwater mussel *Dreissena polymorpha* and found significant levels of TBT bioaccumulated in test organisms; they concluded that larval and juvenile stages may have been affected. Stab *et al.,* (1996) considered the ability for organotin compounds to bioaccumulate through a freshwater food web as "*quite alarming*" and Day *et al.,* (1998) reported potential for toxicity of TBT to some freshwater benthic species from sediments in Canada, both before and after legislation. This serves to highlight that not only marine ecosystems are affected. In developing nations where TBT can still be used on small craft, there is potential for adverse impact on associated freshwater ecosystems.

3.11 TBT and Marine Protected Areas, Crossing Conservation Boundaries

As with atmospheric pollution, marine pollutants have the ability to transcend conservation boundaries (Boersma and Parrish, 1999) and it is imperative that this factor is considered when managing future TBT impacts. Sensitive areas in developing nations have been identified (e.g. Vietnam, Ha Long Bay, UNESCO World Heritage Site) which may be under increasing pressure from marine pollution as economies develop. This situation may be exacerbated by the absence of effective legislation against TBT.

In nations with established economies, the legacy of TBT in sediments is recognised as a continuing problem. TBT can still be used on commercial craft, and once the IMO ban becomes effective there may be a transient increase in the re-charge of TBT levels in sediments, increasing concerns for the safe disposal of butyltin-contaminated material. Anaerobic sediments slow the decomposition of TBT to less toxic chemical species. Estimates of TBT persistence (e.g. 10 years, Dowson *et al.,* 1996 and 20-30 years, Macguire, 2000) and continued occurrence of imposex at sites close to shipping (e.g. Fent, 2004; Santos *et al.,* 2004) indicate the legacy of TBT in sediments could be long-lasting, and appropriate management of potential impacts is an important future consideration.

Internationally, TBT pollution has affected some of the most protected habitats in the world, either directly, from shipping impacts (e.g. Great Barrier Reef, Negri *et al*., 2002; Jones *et al*., 2005), indirectly, from diffuse sources such as sediment (Harino *et al*., 2005[b]) and also from general shipping traffic (Michel, 2001; Terlizzi, 2004). In addition the potential exists to impact protected species, through bioaccumulation (Guruge *et al*., 1996; Strand and Jacobsen, 2005). In other areas (e.g. parts of the south coast of the Isle of Wight) there are apparently anomalous examples where imposex levels are increasing in isolated *N. lapillus* populations with no obvious explanation. As these populations may be resident in Special Areas of Conservation (in this case South Wight) the origins of such abnormal TBT pollution events require further research.

Generally, there is relatively limited information on impacts of TBT pollution within MPAs, PSSAs (Particularly Sensitive Sea Areas. For definition and background see Ünlü, 2004) and other designated protected areas, although much of the wider research into organotins has implications for protected species and habitats. It has been suggested that schemes to create networks of MPAs "*could be compromised by regional and global pollution which easily crosses reserve boundaries*" (Boersma and Parrish, 1999) and clearly such areas are not exempt from wider environmental conditions (Sweeting and Polunin, 2005). In addition, the effectiveness of MPAs is not a global panacea and there are differing regional complexities to consider (see: Sweeting and Polunin, 2005). Thus, TBT inputs from direct (vessels) and indirect (sediment and dockyard practice) sources require proactive management, both in developed and developing nations; even if banned immediately, the legacy of this compound will remain for some years to come.

4. Conclusions

Globally, oceanic TBT pollution was reported to be more evident around Asian waters, based on recent monitoring of skipjack tuna populations. This, together with results of from inshore work, appears to reflect the general concerns of researchers that economic growth in the area, coupled with limited legislation, is resulting in increased marine pollution (including TBT). Organotin impact in sensitive areas around Asia, India and Africa requires further monitoring. In particular, there is limited information for many African ports, whilst trade (and associated shipping movements) close to marine conservation areas in, for example, South Africa is growing. TBT research around Asia appears to be increasing to match increasing trade in the region. This is appropriate, particularly as many coastal human communities depend on mariculture for staple food sources. Future issues of endocrine disrupting chemicals in humans and wildlife will need to focus on developing countries.

In developed nations the problems associated with TBT pollution have been recognised for many years and biological-, water- and sediment-monitoring techniques have been used to demonstrate recovery in many areas in response to legislation. However, as TBT can still be used on commercial vessels, associated hot-spots of contamination have become the focus of much of the current research. These hot-spots are usually linked with large ports, maintenance facilities and, subtidally, with shipping channels, although much of the TBT pollution in the Baltic was found to be derived from recreational craft; there are also concerns about continued illegal use elsewhere. The ability of TBT to remain bound in sediments and the potential for re-release during disturbance is a concern in terms of dredging and disposal of spoil. It has been shown that TBT can be resuspended from dredgings and this may have implications for nearby protected areas, perhaps requiring application of the precautionary principal to ensure safe disposal of spoil.

Marine Protected Areas, PSSAs and other designated conservation sites can only be effective if human operations are managed to minimise impact upon them. Good intentions can be compromised due to the nature of the marine environment; this is not a closed system and pollutants can be transported over considerable distances. With this in mind efforts should be made to ensure the safe disposal of sediments, to consider the nature and frequency of vessel operations near to sensitive areas and, in this context, the continued use of organotin antifouling paints.

The legislation against TBT should be ratified as soon as possible and recent progress suggests that this will happen in the near future. However, as some developing nations may not comply fully with legislation, and as triorganotins can persist for some years, it is reasonable to assume that the legacy of TBT pollution may be with us for a considerable period before concentrations decline, globally, to non-toxic levels.

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Appendix 1

INTERNATIONAL CONVENTION ON THE CONTROL OF HARMFUL ANTI-FOULING SYSTEMS ON SHIPS, 2001
Done at London on 5 October 2001

Entry into force: Not yet in force

Signatories

Australia	"Subject to ratification"
Belgium	"Subject to ratification"
Brazil	"Subject to ratification"
Denmark	"With reservation to Faroe Islands and Greenland"
Finland	"Subject to acceptance"
Morocco	"Subject to ratification"
Sweden	"Subject to ratification"
United States of America	"Subject to ratification

Contracting Governments: as at 16th April 2006

	Date of signature or deposit of instrument	Date of entry into force or succession
Antigua and Barbuda (accession)	6 January 2003	
Bulgaria (accession)	3 December 2004	
Cyprus (accession)	23 December 2005	
Denmark (signature)	19 December 2002	
Greece (accession)	22 December 2005	
Japan (accession)	8 July 2003	
Latvia (accession)	9 December 2003	
Luxembourg (accession)	21 November 2005	
Nigeria (accession)	5 March 2003	
Norway (accession)	5 September 2003	
Poland (accession)	9 August 2004	
Romania (accession)	16 February 2005	
Saint Kitts and Nevis (accession)	30 August 2005	
Spain (accession)	16 February 2004	
Sweden (ratification)	10 December 2003	
Tuvalu (accession)	2 December 2005	

Number of Contracting States: 16, representing approximately 17.27% of the world's merchant shipping

Entry into force

This Convention shall enter into force twelve months after the date on which not less than twenty-five States, the combined merchant fleets of which constitute not less than twenty-five percent of the gross tonnage of the world's merchant shipping, have either signed it without reservation as to ratification, acceptance or approval, or have deposited the requisite instrument of ratification, acceptance, approval or accession in accordance with article 17.